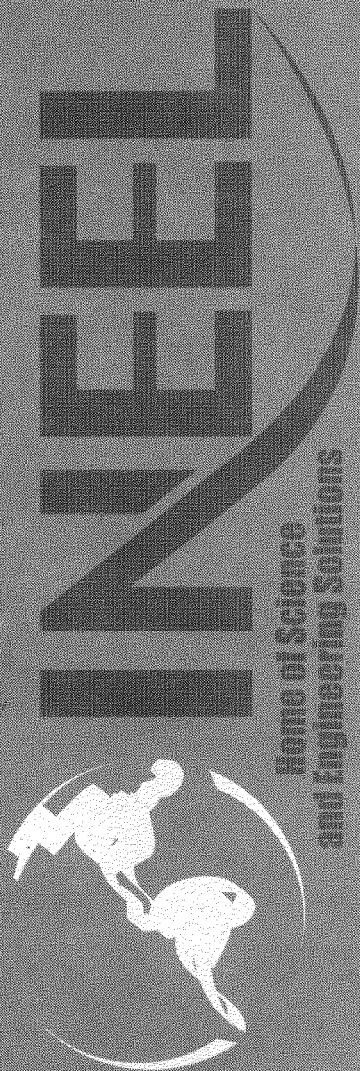


Criticality Safety Evaluation for In Situ Thermal Desorption at the Radioactive Waste Management Complex

Mark Neeley

September 2003



*Idaho National Engineering and Environmental Laboratory
Bechtel BWXT Idaho, LLC*

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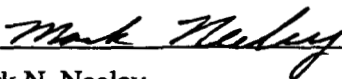
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9/24/03

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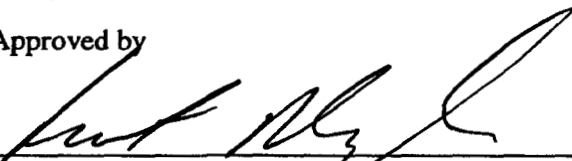


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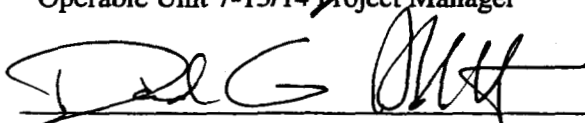
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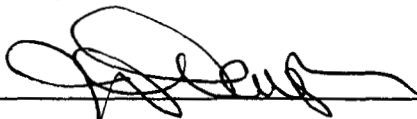
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ABSTRACT

This document addresses the potential for a criticality in the Subsurface Disposal Area (SDA) due to the proposed in situ thermal desorption process. A criticality safety study was performed to address issues relating to postulated criticality scenarios in the SDA for Operable Unit 7-13/14 in the Radioactive Waste Management Complex at the Idaho National Engineering and Environmental Laboratory.

Based on the results of this study, a criticality resulting from the application of the in situ thermal desorption process is not credible with the expected fissile waste forms in the SDA.

EXECUTIVE SUMMARY

The Subsurface Disposal Area (SDA) is a portion of the Radioactive Waste Management Complex at the Idaho National Engineering and Environmental Laboratory that was established in 1952 as a disposal site for solid low-level radioactive waste. Transuranic waste was received from the Rocky Flats Plant and buried in the SDA from 1954 to 1970. This study examines criticality safety issues associated with the use of in situ thermal desorption (ISTD) as a means of removing or destroying hazardous constituents in the buried transuranic waste in the SDA. For this criticality safety evaluation, only ^{239}Pu was analyzed, since it is the most reactive and abundant fissile material reported in the SDA. The ISTD process is a pretreatment to be followed either by retrieval or by in situ grouting of the buried waste. This analysis considers only the ISTD portion of the entire process. Further criticality analysis will be performed to consider the safety of retrieval or in situ grouting if the waste is treated with the ISTD process.

Various configurations were evaluated to determine if any criticality concerns arose in conjunction with treating the buried transuranic waste contained in the SDA with ISTD. This evaluation consisted of three phases. The first phase considered criticality scenarios during the initial application of ISTD. The second phase evaluated the final configurations as a result of applying ISTD. The third phase addressed ancillary issues relating to ISTD and criticality safety.

During the initial application of ISTD, the fissile-bearing waste within the SDA is subjected to various physical and mechanical processes. These processes led to the development of the scenarios and final configurations that were evaluated.

One scenario is fissile material being mixed with plastic and forming an unsafe condition before the ISTD process removes the plastics. Another scenario is the formation of an unsafe condition because of water reentry after the completion of the ISTD process.

Additionally, this evaluation addressed the credibility of forming a critical system in the off-gas collection system. Some of the postulated configurations were evaluated by qualitative means, while other configurations were addressed through computational modeling. Based on the results of this study, a criticality due to the application of the ISTD process is not credible with the expected fissile waste forms in the SDA.

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ACRONYMS

DOE	Department of Energy
HEPA	high-efficiency particulate air (filter)
INEEL	Idaho National Engineering and Environmental Laboratory
ISG	in situ grouting
ISTD	in situ thermal desorption
ISV	in situ vitrification
OU	Operable Unit
RFP	Rocky Flats Plant
RWMC	Radioactive Waste Management Complex
SDA	Subsurface Disposal Area
WAG	Waste Area Group

Criticality Safety Evaluation for In Situ Thermal Desorption at the Radioactive Waste Management Complex

1. INTRODUCTION

The in situ thermal desorption (ISTD) process has been proposed as a means of removing and destroying organic and nitrate contaminants from the soil in the Subsurface Disposal Area (SDA) at the Radioactive Waste Management Complex (RWMC) within the Idaho National Engineering and Environmental Laboratory (INEEL). This evaluation analyzes the criticality safety of the ISTD process using current design information.

2. DESCRIPTION

2.1 Radioactive Waste Management Complex

The RWMC was established in the early 1950s as a disposal site for solid low-level waste generated by INEEL operations. Within the RWMC is the SDA, where radioactive waste materials have been buried in underground pits, trenches, and soil vault rows. Transuranic waste was disposed in the SDA from 1952 to 1970 and was received from Rocky Flats Plant (RFP) for disposal in the SDA from 1954 to 1970. The RFP is a U.S. Department of Energy (DOE)-owned facility located west of Denver, Colorado, and was used primarily for the production of components for nuclear weapons. The RWMC has been designated as Waste Area Group (WAG) 7 and is subdivided into 14 Operable Units (OUs). Technology demonstrations for ISTD will be performed in OU 7-13/14.

The RWMC assigns a content code to each waste container. The content codes are based on the process used for the waste. "Sludges" make up the predominant mass and volume of the waste. There are three general process "sludge" type wastes: inorganic (741-742), organic (743-744), and salt (745). The other waste is generally debris (concrete/asphalt), metal, and trash (combustibles). For criticality safety purposes, these content codes are grouped into eight waste matrices. A waste matrix can cover a range of materials. Table 1 lists the RWMC criticality waste matrix designations, and gives some examples of waste covered by each matrix.

Table 1. List of RWMC waste matrix designations.

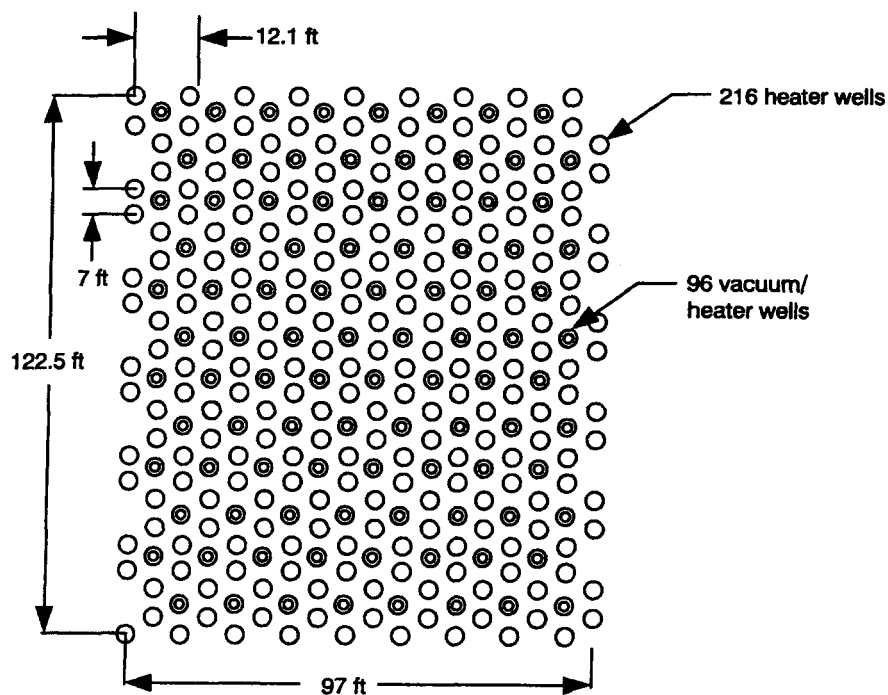
General Classification	Waste Matrix	Examples of Typical Waste
Organic Sludge	Polyethylene	Resins and combustibles
Graphite	Graphite	Graphite molds, heels, and scarfings
Combustible	Cellulose	Benelex, Plexiglas, cemented insulation and filter media
Debris	Brick	Fire brick – scarfed, coarse, pulverized
Debris, Inorganic sludge	Concrete	Cemented and uncemented sludges
Salts	Salt	Evaporated, molten, Gibson, direct oxide reduction salts
Metal	Metal	Noncombustibles, noncompressibles, tantalum, lead
Debris	Glass/slag	Glass bottles, crucibles and molds, dirt, ceramic crucible

2.2 In Situ Thermal Desorption

In situ thermal desorption is a remediation process in which heat and vacuum are applied to contaminated soil. Heat flows into the contaminated soil by conduction from heaters, and gaseous constituents are drawn by vacuum into an off-gas treatment system.

The soil and underground waste are heated using multiple 3-in. schedule 40S stainless steel pipes containing resistive heaters. The heaters are inserted into the waste at 7-ft center-to-center spacing on a triangular pitch. About one-third of the heaters (the middle well of seven in a hexagonal array) are made of slotted 4-in. schedule 40S stainless steel pipe to provide a channel for the gases to be removed by vacuum to an off-gas system. These are called vacuum/heater wells. Figure 1 shows the layout of the wells in an ISTD module. An ISTD module contains 96 vacuum/heater wells, 216 heater wells, and covers approximately 0.27 acres. The heaters drive off water, volatile and semivolatile organic materials. The process also degrades nitrate salts and causes solid organic trash (e.g., paper, plastics, asphalt, cardboard) to combust (in the presence of air) or to pyrolyze (in the absence of air). The fact that each vacuum/heater well contains a heater means that 99% of the organic vapors are destroyed as they pass through the hot soil before they are removed, coming off as carbon dioxide, carbon monoxide, steam, and gaseous hydrochloric acid. Figure 2 is a sketch of a buried vacuum/heater well. Sand between the heater and the slotted vacuum/heater well casing prevents particulate from entering the vacuum/heater well and serves as a roughing filter for the off-gas.

Before treatment, 7 ft of overburden soil will be placed on top of the current 3 ft of overburden, creating 10 ft total of overburden. A moisture barrier will then be placed on top of the soil. Heater wells, vacuum/heater wells, and other subsurface devices will be driven through this overburden, through the waste matrix, through a 2 ft underburden layer, to the basalt below. The waste matrix has an approximate depth of 19 ft. Figure 3 is a sketch of a side view of the waste matrix with inserted ISTD wells.



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Figure 1. Layout of heater wells and vacuum/heater wells in a single ISTD module.

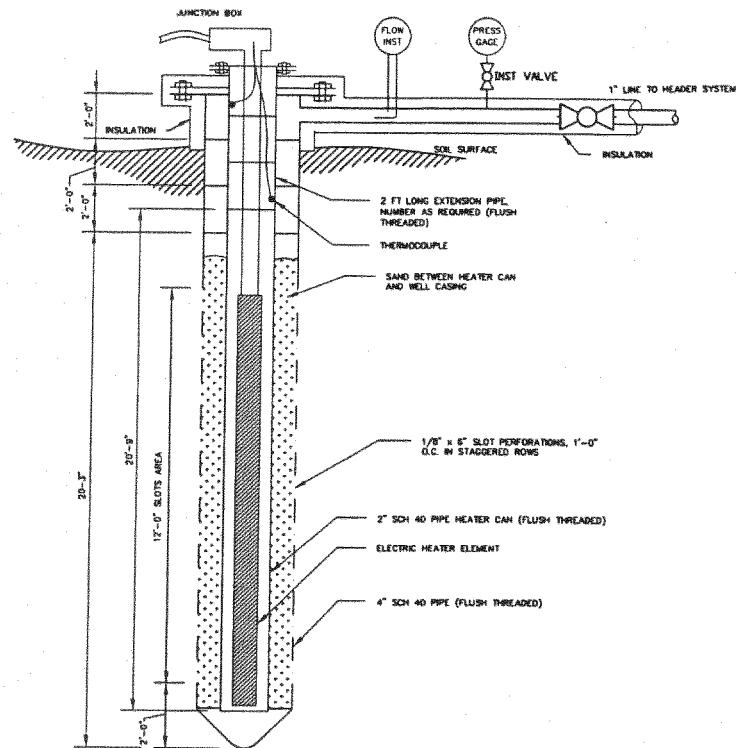


Figure 2. Buried vacuum/heater well.

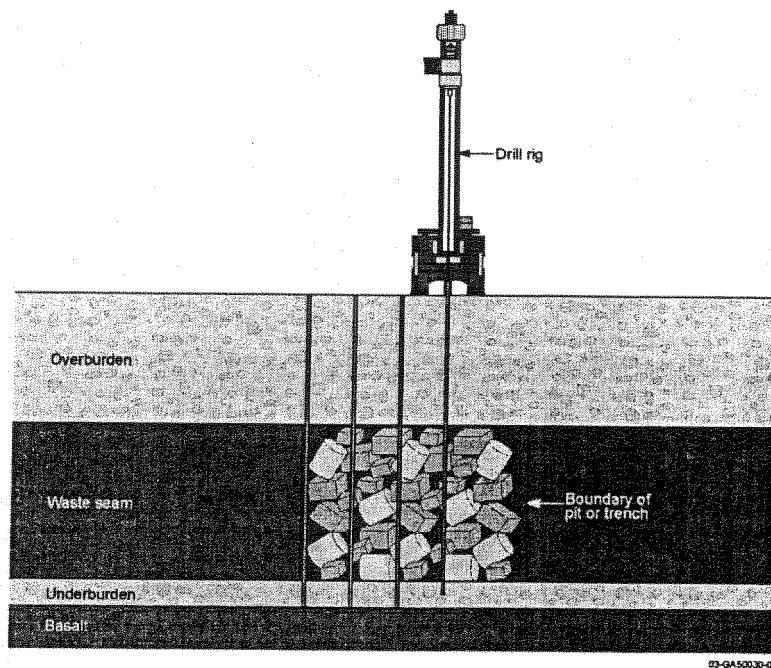


Figure 3. Drill rig used in ISTD process.

During ISTD heating, the temperature gradient rises 2 to 4 C°/cm/day as the water is removed. It stabilizes for up to 2 weeks while water boils off. The temperature gradient then becomes steeper after the water is released, rising at a rate of about 10 to 25 C°/cm/day. Volatile and semivolatile materials decompose or vaporize. Nitrate salts decompose, forming various nitrogen oxide vapors. Organic materials undergo pyrolysis or combustion. At completion of ISTD (estimated at 90 days), the waste volume of material containing organic compounds may be reduced by 60 to 70% depending on the amount of combustibles, soil, the container materials, and the degree of bridging by overlying soil and waste. Although the water, nitrates, and organic contents within the waste drums are removed, the intact drums would still occupy the same volume in the waste seam. The overall effect of ISTD as observed in treating other contaminated soil sites has been a slight subsidence of the waste site of about 2 to 3 in. or about 1% in the affected volume.

The off-gas collection system consists of a cyclone separator, high-efficiency particulate air (HEPA) filters, a regenerative oxidizer, a compact cross-flow heat exchanger, dry gas scrubbers, carbon absorbers, induced draft fans, and an exhaust stack. The cyclone separator removes larger particulates entrained in the off-gas to increase the life of the HEPA filters. The HEPA filters remove radioactive components that may be entrained in the vacuum off-gas, including TRU contaminants of concern (Am-241 and Pu-238, -239, and -240). The thermal oxidizer destroys any halogenated organic compounds that may have been thermally desorbed from the waste stream and did not become oxidized in the subsurface. The off-gas is then cooled through a heat exchanger and passes through a scrubbing unit. The scrubbing unit removes the acid gasses that are present in the off-gas. Following the dry scrubber, the off-gas is passed through carbon bed adsorbers to collect any remaining hydrocarbons. The induced draft fans maintain a negative pressure in the well header piping network and pull the gas stream through the treatment processes and out the exhaust stack.

Further description of the ISTD process can be found in Section 2 of the Feasibility Study – Preliminary Documented Safety Analysis (Abbott 2003).

3. REQUIREMENTS DOCUMENTATION

No unique requirements are applicable to this evaluation. The usual requirements for criticality safety evaluations (e.g. PRD-112, DOE 420.1A) are applicable. However, no limits are developed in this evaluation. This evaluation provides a basis of credibility of postulated criticality scenarios in conjunction with the ISTD process.

4. METHODOLOGY

Calculations were performed using the MCNP, version 4B2, computer code system (RSIC 1997). MCNP is a Monte Carlo transport code used to determine k_{eff} for systems containing fissionable material. The cross-section libraries used for this analysis contained the “point-wise” or “continuous-energy” cross sections.

The analyzed system contained in this report consisted of plutonium dispersed in various waste matrices including water and saturated soil. The geometry of the evaluated systems contained water and plutonium or water saturated soil and plutonium in cylindrical form (drums), spherical form (optimized systems), and rectangular form (infinite slabs). The analyzed systems had H/Pu ratios in the range of 2 to 13.

No critical experiments exist that exactly match the types of systems evaluated. However, modeling critical experiments that encompass the evaluated parameters can validate the various models.

These parameters include material composition, moderation conditions, reflection conditions, and spectral neutron energy ranges.

Validation for these calculations requires experiments that consist of moderated plutonium systems and plutonium combined with silicon.

A validation report was completed that evaluated critical plutonium/silicon configurations (Nielsen 2002). Experiments consisting of plutonium fuel rods, intermixed in a triangular lattice with silicon dioxide rods, were performed in Obninsk, Russia in 1998 and 1999. A detailed description of the critical configurations can be found in (Tsiboulia 2000).

A brief description of the experiments follows. Ten types of rods were used in the plutonium experiments. Each of the rods consisted of a stack of various discs or pellets of various materials. These materials included plutonium metal (canned in stainless steel), silica pellets, polyethylene pellets, stainless steel pellets, and boron carbide pellets. Each of the 10 rods contained a combination of these pellets in a stacked configuration. The rods were then combined to create a critical system. The fuel tubes were arranged in a hexagonal array with a 5.1-cm pitch.

The experiments were modeled as described previously. The calculated results for the experiments, using the ENDF/B-V cross section library, are provided in Table 2. The H/Pu ratio and Si/Pu ratio for the experiments is also presented in the table. The H/Pu ratio varied from 0 to 35 while the Si/Pu ratio varied from 23 to 42. The calculated neutron energy spectrum for these experiments indicates that the energy of the neutrons causing fission is primarily in the intermediate (0.625 eV to 100 keV) to fast (more than 100 keV) range.

Table 2. Polyethylene and silicon dioxide moderated plutonium metal systems.

Case Name	H/Pu	Si/Pu	$k_{eff} \pm \sigma$
BFS-81/1	0	23.4	1.0001 ± 0.0006
BFS-81/1A ^a	0	23.4	0.9987 ± 0.0008
BFS-81/2	2.8	23.4	1.0055 ± 0.0008
BFS-81/3	5.6	23.4	1.0089 ± 0.0008
BFS-81/4	35.2	41.6	1.0178 ± 0.0008
BFS-81/5 ^b	35.2	41.6	1.0164 ± 0.0008
Average: $k_{avg} = \Sigma (k_i/\sigma_i^2) / \Sigma (1/\sigma_i^2)$, $\sigma_{avg} = (1/ \Sigma (1/\sigma_i^2))^{1/2}$ ^c			1.0070 ± 0.0003

a. Differs from Case BFS-81/1 by a different core configuration.
b. Differs from Case BFS-81/5 by a different core configuration.
c. (Bevington 1969).

The last set of cases evaluated consisted of PuO₂/polystyrene cubes reflected by plexiglass plates. Experiments were performed at Hanford between 1963 and 1970 (Justice 2001). Twenty-nine experiments were performed with various configurations, concentrations of plutonium, and plutonium enrichments. The cubes were approximately 2 × 2 × 2 in. and were stacked on a split-table critical assembly. The two halves of the assembly were brought together and the neutron multiplication was determined using proportional counters. Some of the cubes were cut in the axial direction to allow flexibility in obtaining a critical height. The final critical configuration consisted of a rectangular block of PuO₂/polystyrene reflected on all six sides by plexiglass. The H/Pu ratios ranged from 6 to 65 and the C/Pu ratios varied from 6 to 64. A more detailed description of these experiments can be found in an

internal report (Nielsen 2003) that discusses validation of calculations containing Pu/polystyrene. The results from these cases are shown in Table 3. As shown by the results of the validation experiments, no bias caused by calculational methodology is warranted.

Table 3. Polystyrene-moderated plutonium oxide systems reflected by plexiglass.

Case Name	Pu density (g/cm ³)	Wt% Pu-240	H/Pu	C/Pu	Length (cm)	Width (cm)	Height (cm)	$k_{eff} \pm \sigma$
Case 6	2.302	11.46	5.87	5.86	25.60	25.60	18.33	1.0170 \pm 0.0009
Case 7	2.302	11.46	5.87	5.86	30.72	30.72	14.18	1.0177 \pm 0.0008
Case 8	2.302	11.46	5.87	5.86	40.96	40.96	10.59	1.0173 \pm 0.0007
Case 9	2.302	11.46	5.87	5.86	51.20	51.20	9.04	1.0193 \pm 0.0008
Case 10	1.120	2.20	15.5	15.5	51.69	46.13	9.04	1.0285 \pm 0.0010
Case 11	1.120	2.20	15.5	15.5	41.35	38.46	10.34	1.0270 \pm 0.0010
Case 12	1.120	2.20	15.5	15.5	31.01	31.01	13.13	1.0247 \pm 0.0010
Case 13	1.120	2.20	15.5	15.5	25.86	25.86	16.43	1.0233 \pm 0.0009
Case 14	1.120	2.20	15.5	15.5	23.27	23.27	19.79	1.0275 \pm 0.0010
Case 15	1.120	2.20	15.5	15.5	20.68	20.68	24.87	1.0256 \pm 0.0009
Case 16	1.120	2.20	15.5	15.5	15.52	18.08	50.04	1.0214 \pm 0.0010
Case 17	1.050	8.06	16.4	16.4	51.31	68.25	10.36	1.0045 \pm 0.0009
Case 18	1.050	8.06	16.4	16.4	35.92	35.92	15.42	1.0088 \pm 0.0008
Case 19	1.050	8.06	16.4	16.4	30.78	30.78	18.56	1.0051 \pm 0.0007
Case 20	1.050	8.06	16.4	16.4	25.65	25.65	25.03	1.0056 \pm 0.0008
Case 21	1.050	8.06	16.4	16.4	25.65	25.65	25.13	1.0072 \pm 0.0009
Case 22	1.050	8.06	16.4	16.4	20.52	20.52	49.15	1.0101 \pm 0.0008
Case 23	0.367	18.35	65.4	64.4	61.08	61.08	16.35	1.0054 \pm 0.0009
Case 24	0.367	18.35	65.4	64.4	50.90	61.08	17.48	1.0054 \pm 0.0008
Case 25	0.367	18.35	65.4	64.4	50.90	50.90	18.68	1.0069 \pm 0.0017
Case 26	0.367	18.35	65.4	64.4	50.90	45.81	19.69	1.0081 \pm 0.0009
Case 27	0.367	18.35	65.4	64.4	40.72	45.81	22.06	1.0086 \pm 0.0008
Case 28	0.367	18.35	65.4	64.4	40.72	40.72	23.58	1.0091 \pm 0.0009
Case 29	0.367	18.35	65.4	64.4	40.72	30.54	29.64	1.0110 \pm 0.0010
Average: $k_{avg} = \Sigma (k_i/\sigma_i^2) / \Sigma (1/\sigma_i^2)$, $\sigma_{avg} = (1/\Sigma (1/\sigma_i^2))^{1/2}$ ^a								1.0139 \pm 0.0002
a. (Bevington 1969)								

5. DISCUSSION OF CONTINGENCIES

Contingencies are events that may occur, but are not likely or are unintended. Contingencies usually include such events as flooding, placing too much fissile material in a container, or an accident in which fissile fuel is further concentrated from its original form. Possible contingencies were identified as part of this analysis. The evaluations performed in Section 6 assess the various contingencies considered, and show their effect on the criticality safety of the system. The analysis shows that the ISTD process will not cause an inadvertent criticality in the SDA due to any contingency.

To create a critical configuration with reasonable quantities of fissile material, various factors must be met. An unsafe mass of fissile material must be present. This fissile mass must be concentrated, optimally moderated, and in a favorable or optimal geometrical configuration. The system also needs near full reflection and must be free from diluents or neutron-absorbing materials.

The majority of fissile material in the SDA is dispersed at relatively low concentrations. If an area of highly concentrated fissile material exists, the various factors above would need to be near optimal to achieve an unsafe condition. For example, more than 10.2 kg of moist (1.5 wt% water) PuO_2 is required to create an unsafe condition. This system consists of uniform plutonium oxide powder in a small volume, which is free of diluent materials and fully reflected by an infinite perfect reflector. These ideal conditions do not exist in the SDA, and the application of the ISTD process will not create them.

For lower fissile masses and concentrations, the optimal conditions are even more necessary to create an unsafe condition. At low masses, intimate mixing with a moderating material such as water or polyethylene is required to cause a criticality. Water may be introduced into the SDA following the ISTD process, but there is no mechanism available to mix the water and any fissile material that may be present to the extent needed to form a critical configuration. The formation of a critical system is therefore less likely for a system with low fissile mass than with high fissile mass. Therefore, since the idealized conditions that must be met for a criticality to occur do not exist in the SDA, and cannot be created by the ISTD process, an inadvertent criticality is not possible.

6. EVALUATION AND RESULTS

6.1 Criticality Safety Background Information and Definitions

Waste at the RWMC contains fissile isotopes. The fissile isotopes of concern are ^{239}Pu from weapons-grade plutonium and ^{235}U from highly enriched uranium. Much of the fissile material is in oxide, nitrate, or hydrated-oxide form. Criticality is difficult to obtain and requires special conditions and materials. It may be defined as "the attainment of conditions such that fissile material will sustain a chain reaction (Cember 1985)." Criticality is determined by reactivity. The term "reactivity" refers to the deviation of a system from a critical value of $k_{\text{eff}} = 1$, where k_{eff} is the neutron multiplication factor for a system. A more reactive system has greater neutron multiplication. If the neutron multiplication is such that the system is just self-sustaining, it is critical. A supercritical system has a value of k_{eff} greater than 1, while a subcritical system has a k_{eff} value less than 1. The factors that affect reactivity are reflection, moderation, geometry, fissile mass, and fissile mass concentration.

A reflector surrounds the fissile material and reflects neutrons back into the fissile region. Reflectors include water, paraffin, beryllium, graphite, concrete, and thick metal. A moderator is usually intimately mixed with the fissile material. It slows neutrons so that they are more likely to react with the fissile isotopes and cause fission. Many reflectors are also good moderators. Hydrogen is the most common and effective moderator, particularly as found in water, polyethylene, or oils. Hydrogen is an

effective moderating atom because it has almost the same mass as a neutron, enabling almost complete momentum transfer in a single collision. Oil and polyethylene are better moderators than water because they have greater hydrogen density. Beryllium and graphite are also good moderators, but require larger volumes for criticality. Beryllium is actually more effective as a reflector than as a moderator. Each system has an optimally reflected, moderated state where its reactivity is greatest.

The geometry of the fissile material affects the system reactivity. As the ratio of volume to surface area increases for a given volume, neutron leakage decreases and reactivity increases (Paxton and Pruvost 1986). The optimum geometry is thus usually a sphere that has maximum volume and minimum surface area. The type of fissile isotope affects the system reactivity. Odd-numbered isotopes usually fission due to the absorption of thermal neutrons more readily than even numbered isotopes. Even-numbered isotopes more readily fission due to fast spectral neutrons. Thus, ^{235}U and ^{239}Pu are much more fissionable than ^{238}U and ^{240}Pu . The concentration of the fissile isotope is important to the system reactivity. If the fissile material is too dilute, criticality cannot be achieved.

Moderation and reflection usually work together since most moderators also act as reflectors. A fissile material solution in a moderator, such as water, has a smaller critical mass. The minimum critical mass in water for each of the fissile isotopes corresponds to an optimally moderated and homogeneous mixture of the metal isotope and water in a spherical configuration with full water reflection. These values are specified as the "solution critical mass." The critical mass of the solid metal is more than ten times that for the isotope in solution. For example, the critical mass of solid plutonium metal is about 6 kg, whereas the critical mass of plutonium in solution is about 0.5 kg. Moist-oxide critical masses are significantly greater (about 10 kg for moist PuO_2) than those for solutions and metals. Deviations from the ideal conditions increase the critical mass.

For criticality to occur in the SDA (due to the ISTD process), several unlikely concurrent parameters must exist: (a) there must be sufficient fissile mass, (b) the fissile mass must be at or near the optimum concentration, (c) the fissile mass must be in a near-optimal geometry, (d) near-optimal reflection must exist, and (e) the fissile mass must be in a waste matrix that lacks diluents and neutron absorbers. The ISTD process will remove existing moderating materials from the matrix, but does not preclude moderator from seeping back into the matrix.

6.2 Analysis Overview

Various configurations were evaluated to determine if any criticality concerns arise in conjunction with using the ISTD process to treat buried waste contained in the SDA. This evaluation consists of three phases: (1) considering criticality scenarios during the initial application of the ISTD process, (2) evaluating the final configurations as a result of the application of the ISTD process, and (3) addressing ancillary issues relating to ISTD and criticality safety. Each of these phases is further described in the remainder of this section.

During the initial application of the ISTD process, the fissile-bearing waste within the SDA is subjected to various processes. These processes include the melting of plastics that could entrain fissile material, and subsidence in the waste zone. Plutonium and uranium are not soluble in nitrate salts (Morgan 1980), so entrainment in molten nitrates is not a criticality concern.

This evaluation will address the effects of water reentry into the final configuration, and whether it is credible to form a critically unsafe condition in the off-gas collection system.

Some of the postulated configurations were evaluated qualitatively while other configurations were addressed through computational modeling. For this criticality safety evaluation, only ^{239}Pu was analyzed since it is the most reactive and abundant fissile material reported in the SDA.

6.3 Concentration of Plutonium or Uranium Metal

One postulated scenario of concern is concentration of plutonium or uranium metal during the application of the ISTD process and the formation of a critical configuration. The ISTD process will cause volume reduction in the waste matrix, and will therefore have a tendency to concentrate the fissile material.

Some plutonium might have originally been deposited in metallic form, specifically that associated with metal crucibles (metal waste matrix) and nonmetal molds and crucibles (graphite, glass/slag waste matrices). Any large piece of plutonium or uranium in metallic form is expected to at least have an outer oxide film. Small metal pieces are expected to be completely oxidized (ANS 1980).

Plutonium readily oxidizes in air. Calculations of oxidation rates for ^{239}Pu metal as a single sphere surrounded by water, show that after 27 years a 0.5 in. diameter sphere containing 22 g ^{239}Pu would be completely oxidized (Sentieri 2003). The assumption of a spherical shape with minimum surface area is overly conservative. Flat shapes have more surface area, division of the material into more than one piece increases surface area, and most of the material was in the form of contamination (i.e. finely divided). ISTD will not reduce oxides back to a metal. The oxide is very stable.

The oxidation potentials for uranium and plutonium are sufficiently high that ISTD would not result in reduction of the oxides to the metal. The melting temperature of PuO_2 is $2,400^\circ\text{C}$ and the maximum temperatures expected in ISTD are about 800°C . PuO_2 can be reduced in the presence of tantalum or calcium to form a slag. However, the process is not very effective. In addition, there is no tantalum in the soil at INEEL and calcium is present only as the oxide. There is actually a good amount of CaO in the soil, but the fact that it is already oxidized means it does not seek after the oxygen from PuO_2 . Therefore, the driving force for reduction of PuO_2 to metal is not in the waste.

The oxide is generally an insoluble form. No credible concentration mechanism for a large amount of fissile material has been identified. Since plutonium and uranium are not soluble in nitrate salts (Morgan 1980), the fissile material will not be entrained in the molten nitrates and will not preferentially collect in one place due to the nitrates. The mixing of fissile material with organic materials such as polyethylene is addressed in Section 6.4.

The single parameter limit for ^{239}Pu in a moist-oxide form (1.5 wt% H_2O), rather than a solution, is 10.2 kg (ANSI 8.1) for a system at full density and fully reflected by water. The total quantity of fissile isotopes buried at the SDA has been estimated to be about 350 kg of actinides. However, the fissile material is mostly dispersed at low concentration throughout the waste (Sentieri 2003). Fissile material exists primarily as contamination on the waste material. A few items may potentially contain larger amounts of fissile material, such as filters and graphite material. These materials make up a small percentage of the total waste material both by mass and volume (Clements 1982).

The low overall concentrations of fissile material within the waste, in conjunction with the high probability that oxidation of any metal has occurred, and the difficulty in reducing oxide to metal form due to ISTD, lead to the conclusion that the formation of a critical system due to the concentration of plutonium or uranium metal within the waste matrices of the SDA is not credible.

6.4 Mixtures of Fissile Material and Polyethylene

The next set of postulated configurations considers the combination of fissile material and polyethylene. Only plutonium was considered since it is the most reactive and abundant fissile material reported in the SDA. During the ISTD process, the temperatures are high enough to drive off organic compounds such as polyethylene, as well as any other liquids. Most of the waste contained in the soil will melt and/or vaporize at their respective melting and vaporization points. Out of the present waste matrices, the most reactive waste form would be polyethylene when combined with plutonium.

In the case of polyethylene, several things must happen to cause a criticality during heating of buried waste. Polyethylene first must melt during the initial phase of ISTD before temperatures reach levels sufficient to destroy it. The melted polyethylene must selectively entrain or combine in a homogeneous fashion with the fissile isotopes (^{239}Pu or ^{235}U). The melted plastic and fissile material must then flow and concentrate in a single area, or from one area to another. For reasonable fissile masses, this arrangement must be of sufficient concentration and proper shape to optimally moderate neutrons to cause a criticality.

Polyethylene, cellulose, and graphite are present in some waste matrices, and represent effective carbon-based neutron moderators and reflectors (Paxton and Pruvost 1986). Polyethylene is superior to water as a neutron reflector/moderator. Polyethylene is a thermoplastic, which melts at 85 to 110°C. The exact temperature varies with physical properties such as the density, cross-linking frequency, and the degree of crystallinity. Cellulose will decompose at 260 to 270°C, rather than melt (CRC 1982). Graphite, also an effective moderator, does not melt or decompose, but reacts with oxygen at 110°C (CRC 1982). Virtually all moderators (except graphite), including water and most organic materials, leave the heated area undergoing volatilization and destruction (combustion, if oxidizer is present; or pyrolysis, if oxidizer is absent). Polyethylene plastic begins melting with the water vaporization and is completely melted after the water is gone. The moderating water will not be present by the time polyethylene has had sufficient time to melt and pool. Polyethylene is not likely to concentrate fissile material to any extent because it will continue to flow until it pyrolyzes or volatilizes. The solubility of fissile materials in molten polyethylene plastics is likely to be very low, based on the insolubility of most metals and oxides, including Pu, in aliphatic nonpolar organic materials. Polyethylene, even in larger quantities, does not have the ability to entrain or dissolve appreciable amounts of fissile material, nor does it have any concentrating capacity.

Polyethylene is very viscous during a slow melt. The speed of the heating would determine whether the polyethylene would melt and flow before it is vaporized or pyrolyzed. Polyethylene fluidity in the temperature range between melting and decomposition is low. Although there could be localized movement of molten polyethylene, there will be little if any movement within the waste (a must for postulating the sufficient concentration of fissile material). Moderation from this material is thus not credible for multiple containers on a pit-wide basis.

Calculations performed in (Sentieri 2003) demonstrate the fissile masses necessary to postulate a critical system composed of plutonium and polyethylene, in conjunction with the optimal geometry, reflection conditions, fissile concentration, and lack of diluent/absorber material. The amount of fissile mass necessary in a localized area, and the concurrent conditions necessary, lead to the conclusion that the formation of a critical system resulting from the initial application of the ISTD process is not credible. There is no conceivable mechanism to mix the plutonium homogeneously in the molten polyethylene at the near-optimum concentration required for a criticality.

6.5 Flooding and/or Water Reentry

A concern to be addressed is the chance of a criticality occurring if water percolates back into the waste matrix following the ISTD process. Voids could exist in the waste matrix after the ISTD process, but the process will not cause voids in any fissile oxides that may be present in the waste matrix. The analysis in this section shows that there would not be sufficient void volume in the fissile oxide such that it could become intimately mixed with water on the scale needed to cause a criticality. The SDA does not lie on a flood plain; however, on at least three occasions local runoff from rapid spring thaws caused flooding that covered part of the SDA with water (Holdren et al. 2002). A 4.6-m (15-ft) dike has since been constructed around the SDA to prevent future flooding. The SDA is exposed to the elements, and there is a potential for water intrusion.

Flooding while the wells are being placed in the ground is not considered to be a criticality concern. The analysis of the waste matrix in its current form shows there is no criticality concern due to flooding (Sentieri and Taylor 2003). The drilling of the wells will not change the form of the waste matrix enough to invalidate the conclusion of that analysis. Current requirements for coring and probing in the SDA state that probe casings and core holes shall have a maximum 6-in. internal diameter, and that there shall be a minimum 5-ft edge-to-edge distance between the probe casings or core holes. The design of the ISTD wells satisfies both of these requirements, since the wells have a maximum 4-in. nominal inside diameter, and have a nominal 6-ft, 7.5-in. edge-to-edge space between the wells. In addition, a moisture barrier will be placed on top of the soil before the ISTD wells are drilled. This barrier will help ensure that water will not enter the soil during the ISTD process.

During ISTD, plutonium and uranium oxides remain in their oxide form. The ISTD process will combust or pyrolyze much of the contents of the waste drums. The remaining contents will then collect at the bottom of the drums. Since fissile materials are not expected to be destroyed by the ISTD process, they will also collect at the bottom of the drum. This is not a criticality concern since it takes about 6 kg of solid plutonium metal or more than 10 kg of PuO_2 to cause a criticality. The addition of water via a flooding event after the completion of the ISTD process could increase the reactivity of the SDA, but not enough to be a criticality concern. The reintroduction of water can only cause concern if it can dissolve the fissile material, collect it in one place, and then become intimately mixed with the fissile material in an optimally moderated, homogeneous mixture. The fissile metals and oxides are stable, and will not dissolve in water. Only gram quantities of fissile material are expected to be in the waste, and are near insoluble in water. Water in the waste following treatment must have intimate contact with the fissile material on an atomic scale (as in a solution) to be effective as a moderator. There is no mechanism inside the SDA that will cause fissile material to form an optimally moderated homogeneous mixture with water.

Movement of soil into voids left by water and organic materials will reduce the opportunity for these moderators to return. The overall effect of ISTD as observed in treating other contaminated soil sites has been a slight subsidence of the waste site of about 2 to 3 in. or about 1% in the affected volume. The application of the ISG process to an area that has undergone ISTD will add moderator in the form of wet cementitious grout. A separate analysis will be performed that considers the criticality safety of performing ISG on an area that has been previously treated by the ISTD process.

A criticality analysis of in situ vitrification (ISV) of Pu in INEEL soil can be used for ISTD (Sentieri 2003). For PuO_2 and for oxide mixed with representative INEEL soil, theoretical densities of 11.46 g/cm^3 for the oxide and 2.38 g/cm^3 for the compacted soil were assigned. These densities were therefore used in the subsequent analysis.

The waste in the SDA is assumed to be critically safe in its current configuration (Sentieri and Taylor 2003). The analysis of plutonium metal oxidation before any thermal treatment demonstrates that

at room temperature and 100% relative humidity, spherical particles less than 0.5 in. in diameter will completely oxidize in 27 years (Sentieri 2003). All waste below ground in the WAG 7 waste pits has been buried for at least that long. It is reasonable to expect that the waste has been subject to high relative humidity at times, especially with at least three flood events during the recorded history of the SDA (Holdren et al. 2002). Uranium metal is expected to oxidize in a similar fashion. If the oxidation of Pu is not complete because of organic coatings, it will be oxidized during ISTD heating, as these coatings are removed.

There are three possible forms for plutonium and uranium at the start of processing: oxide, salt, and metal. Pu and U are thermodynamically stable in the oxide form. The fissile oxides are nonvolatile, with low vapor pressures and melting points greater than the ISTD temperatures. The heating proceeds slowly, and the vacuum does bring in small amounts of air so oxides at a particular location have adequate time to form.

Plutonium and uranium salts, particularly nitrates, might dissociate as temperature rises. The positively charged fissile ions in this case will oxidize. The temperature is insufficient to vaporize halide, carbonate, and sulfate salts.

Cases were analyzed in which various shapes of water-moderated plutonium dioxide were surrounded by water-saturated soil. Again, only plutonium was considered since it is the most reactive and abundant fissile material reported in the SDA. The shapes considered were spheres, slabs having the same diameter (22.5 in.) as a 55-gal drum, and infinite slabs. The plutonium dioxide concentration was varied to find the critical PuO_2 mass or dimension for the various concentrations and shapes considered. Plutonium dioxide was considered because it is more reactive than uranium, and will therefore envelope the results for uranium. The PuO_2 concentrations considered represent between 60% (4.584 g/cm^3) and 83% (1.948 g/cm^3) porous plutonium dioxide. In reality, bulk plutonium dioxide ranges in density from about 4.5 g/cm^3 to 6.5 g/cm^3 (ANS 1980). The void spaces in the PuO_2 were filled either with water or water saturated soil. Results of the analysis are shown in Tables 4 through 9, and are summarized in Table 10.

The cases in Tables 4 through 9 were analyzed to show the mass and dimensions necessary to cause a criticality in the SDA due to water intrusion after ISTD treatment. The results show in general that as concentration decreases, the critical mass of plutonium dioxide decreases. The fissile concentrations were not further decreased since there is no credible mechanism to mix water and fissile material at any lower concentrations than those considered. Calculations for infinite slabs (Tables 8 and 9) were performed to show the effects of having an infinite array of drums each containing water-moderated plutonium dioxide. The results show that it would take about 8.4 kg of porous plutonium dioxide (the void space filled with water) to cause a criticality, or about 22.6 kg with the void space filled with water saturated soil. It is not credible that this much fissile material will collect in one location. The results also show that a 2.8-cm-thick infinite slab would cause a criticality, but additional calculations in Appendix C show that 700 kg of plutonium dioxide would be needed to fill this slab, which is double the estimated amount of total actinides in the entire SDA. It is not credible that this amount of fissile material could preferentially collect to form a critical system.

Table 4. Sphere of water-saturated PuO₂, surrounded by water-saturated soil.

Case Name	Concentration (g PuO ₂ /cm ³)	H/X	Sphere Radius (cm)	PuO ₂ Mass in Sphere (kg)	$k_{\text{eff}} \pm 1\sigma$	$k_{\text{eff}} + 2\sigma$
TD066	4.584	3.9	7.5	8.10	0.8672 ± 0.0018	0.8708
TD062	4.584	3.9	8.0	9.83	0.9191 ± 0.0017	0.9225
TD067	4.584	3.9	8.5	11.79	0.9681 ± 0.0019	0.9719
TD063	4.584	3.9	9.0	14.00	1.0145 ± 0.0019	1.0183
TD076	4.000	4.9	7.5	7.07	0.8474 ± 0.0016	0.8506
TD073	4.000	4.9	8.0	8.58	0.8984 ± 0.0020	0.9024
TD077	4.000	4.9	8.5	10.29	0.9467 ± 0.0020	0.9507
TD074	4.000	4.9	9.0	12.21	0.9908 ± 0.0018	0.9944
TD071	4.000	4.9	9.5	14.37	1.0395 ± 0.0018	1.0431
TD083	3.438	6.1	8.0	7.37	0.8772 ± 0.0019	0.8810
TD087	3.438	6.1	8.5	8.84	0.9306 ± 0.0018	0.9342
TD084	3.438	6.1	9.0	10.50	0.9728 ± 0.0019	0.9766
TD086	3.438	6.1	9.5	12.35	1.0122 ± 0.0017	1.0156
TD093	2.865	7.9	8.0	6.14	0.8541 ± 0.0020	0.8581
TD097	2.865	7.9	8.5	7.37	0.9033 ± 0.0018	0.9069
TD094	2.865	7.9	9.0	8.75	0.9501 ± 0.0018	0.9537
TD096	2.865	7.9	9.5	10.29	0.9922 ± 0.0019	0.9960
TD092	2.865	7.9	10.0	12.00	1.0324 ± 0.0020	1.0364
TD107	2.292	10.5	8.5	5.90	0.8793 ± 0.0019	0.8831
TD104	2.292	10.5	9.0	7.00	0.9255 ± 0.0019	0.9293
TD106	2.292	10.5	9.5	8.23	0.9637 ± 0.0017	0.9671
TD102	2.292	10.5	10.0	9.60	1.0078 ± 0.0018	1.0114
TD113	1.948	12.8	8.5	5.01	0.8627 ± 0.0019	0.8665
TD114	1.948	12.8	9.0	5.95	0.9118 ± 0.0018	0.9154
TD117	1.948	12.8	9.5	7.00	0.9528 ± 0.0021	0.9570
TD112	1.948	12.8	10.0	8.16	0.9887 ± 0.0015	0.9917
TD118	1.948	12.8	10.5	9.46	1.0256 ± 0.0019	1.0294

Table 5. Sphere of PuO₂ and water-saturated soil, surrounded by water-saturated soil.

Case Name	Concentration (g PuO ₂ /cm ³)	H/X	Sphere Radius (cm)	PuO ₂ Mass in Sphere (kg)	$k_{\text{eff}} \pm 1\sigma$	$k_{\text{eff}} + 2\sigma$
TD202	4.584	1.6	9.0	14.00	0.8741 ± 0.0016	0.8774
TD201	4.584	1.6	9.5	16.46	0.9143 ± 0.0017	0.9177
TD205	4.584	1.6	10.0	19.20	0.9520 ± 0.0017	0.9554
TD204	4.584	1.6	10.5	22.23	0.9923 ± 0.0018	0.9959
TD203	4.584	1.6	11.0	25.56	1.0303 ± 0.0017	1.0337
TD211	4.000	2.0	9.5	14.37	0.8716 ± 0.0018	0.8752
TD215	4.000	2.0	10.0	16.76	0.9088 ± 0.0017	0.9122
TD214	4.000	2.0	10.5	19.40	0.9484 ± 0.0018	0.9520
TD213	4.000	2.0	11.0	22.30	0.9853 ± 0.0016	0.9889
TD212	4.000	2.0	11.5	25.48	1.0204 ± 0.0018	1.0240
TD225	3.438	2.5	10.0	14.40	0.8688 ± 0.0016	0.8720
TD224	3.438	2.5	10.5	16.67	0.9048 ± 0.0019	0.9086
TD223	3.438	2.5	11.0	19.17	0.9409 ± 0.0019	0.9447
TD222	3.438	2.5	11.5	21.90	0.9723 ± 0.0017	0.9757
TD221	3.438	2.5	12.0	24.89	1.0064 ± 0.0018	1.0100
TD232	2.865	3.2	11.0	15.97	0.8917 ± 0.0017	0.8951
TD233	2.865	3.2	11.5	18.25	0.9253 ± 0.0018	0.9289
TD235	2.865	3.2	12.0	20.74	0.9563 ± 0.0019	0.9601
TD234	2.865	3.2	12.5	23.44	0.9895 ± 0.0019	0.9933
TD231	2.865	3.2	13.0	26.37	1.0202 ± 0.0018	1.0238
TD246	2.292	4.2	11.5	14.60	0.8762 ± 0.0017	0.8796
TD245	2.292	4.2	12.0	16.59	0.9072 ± 0.0018	0.9108
TD244	2.292	4.2	12.5	18.75	0.9380 ± 0.0019	0.9418
TD241	2.292	4.2	13.0	21.09	0.9676 ± 0.0018	0.9712
TD243	2.292	4.2	13.5	23.62	0.9944 ± 0.0018	0.9980
TD242	2.292	4.2	14.0	26.34	1.0224 ± 0.0017	1.0258
TD257	1.948	5.1	12.0	14.10	0.8732 ± 0.0017	0.8766
TD255	1.948	5.1	12.5	15.94	0.9087 ± 0.0019	0.9125
TD251	1.948	5.1	13.0	17.93	0.9311 ± 0.0017	0.9345
TD254	1.948	5.1	13.5	20.08	0.9603 ± 0.0017	0.9637
TD253	1.948	5.1	14.0	22.39	0.9865 ± 0.0018	0.9901
TD256	1.948	5.1	14.5	24.88	1.0141 ± 0.0018	1.0177

Table 6. Slab of water-saturated PuO₂ in 55-gallon drum, surrounded by water-saturated soil.

Case Name	Concentration (g PuO ₂ /cm ³)	H/X	Slab Thickness (cm)	PuO ₂ Mass in Cylinder (kg)	$k_{\text{eff}} \pm 1\sigma$	$k_{\text{eff}} + 2\sigma$
TD135	4.584	3.9	2.0	23.52	0.8429 ± 0.0018	0.8465
TD134	4.584	3.9	2.5	29.40	0.8996 ± 0.0016	0.9028
TD131	4.584	3.9	3.0	35.28	0.9543 ± 0.0018	0.9579
TD133	4.584	3.9	3.5	41.16	1.0026 ± 0.0017	1.0060
TD144	4.000	4.9	2.5	25.65	0.8857 ± 0.0018	0.8893
TD141	4.000	4.9	3.0	30.78	0.9374 ± 0.0017	0.9408
TD143	4.000	4.9	3.5	35.91	0.9889 ± 0.0018	0.9925
TD142	4.000	4.9	4.0	41.04	1.0292 ± 0.0019	1.0330
TD154	3.438	6.1	2.5	22.05	0.8688 ± 0.0018	0.8724
TD151	3.438	6.1	3.0	26.46	0.9195 ± 0.0018	0.9231
TD153	3.438	6.1	3.5	30.87	0.9649 ± 0.0018	0.9685
TD152	3.438	6.1	4.0	35.28	1.0072 ± 0.0017	1.0106
TD164	2.865	7.9	2.5	18.37	0.8505 ± 0.0017	0.8539
TD161	2.865	7.9	3.0	22.05	0.9021 ± 0.0018	0.9057
TD163	2.865	7.9	3.5	25.72	0.9465 ± 0.0016	0.9497
TD162	2.865	7.9	4.0	29.40	0.9890 ± 0.0018	0.9926
TD165	2.865	7.9	4.5	33.07	1.0274 ± 0.0017	1.0303
TD171	2.292	10.5	3.0	17.64	0.8773 ± 0.0018	0.8809
TD173	2.292	10.5	3.5	20.58	0.9239 ± 0.0017	0.9273
TD172	2.292	10.5	4.0	23.52	0.9671 ± 0.0017	0.9705
TD174	2.292	10.5	4.5	26.46	1.0035 ± 0.0019	1.0073
TD185	1.948	12.8	3.0	14.99	0.8637 ± 0.0017	0.8671
TD181	1.948	12.8	3.5	17.49	0.9102 ± 0.0019	0.9140
TD183	1.948	12.8	4.0	19.99	0.9517 ± 0.0017	0.9551
TD182	1.948	12.8	4.5	22.49	0.9862 ± 0.0017	0.9896
TD184	1.948	12.8	5.0	24.99	1.0236 ± 0.0019	1.0274

Table 7. Slab of PuO₂ and water-saturated soil in 55-gallon drum, surrounded by water-saturated soil.

Case Name	Concentration (g PuO ₂ /cm ³)	H/X	Slab Thickness (cm)	PuO ₂ Mass in Cylinder (kg)	$k_{\text{eff}} \pm 1\sigma$	$k_{\text{eff}} + 2\sigma$
TD321	4.584	1.6	3.0	35.28	0.8762 ± 0.0016	0.8794
TD324	4.584	1.6	3.5	41.16	0.9164 ± 0.0018	0.9200
TD322	4.584	1.6	4.0	47.04	0.9569 ± 0.0018	0.9605
TD325	4.584	1.6	4.5	52.92	0.9919 ± 0.0014	0.9947
TD323	4.584	1.6	5.0	58.79	1.0275 ± 0.0018	1.0311
TD334	4.000	2.0	3.5	35.91	0.8880 ± 0.0017	0.8914
TD332	4.000	2.0	4.0	41.04	0.9271 ± 0.0018	0.9307
TD335	4.000	2.0	4.5	46.17	0.9599 ± 0.0020	0.9639
TD333	4.000	2.0	5.0	51.30	0.9921 ± 0.0017	0.9955
TD336	4.000	2.0	5.5	56.43	1.0239 ± 0.0016	1.0271
TD342	3.438	2.5	4.0	35.28	0.8961 ± 0.0017	0.8995
TD345	3.438	2.5	4.5	39.69	0.9259 ± 0.0016	0.9291
TD343	3.438	2.5	5.0	44.10	0.9584 ± 0.0018	0.9620
TD346	3.438	2.5	5.5	48.51	0.9845 ± 0.0018	0.9881
TD344	3.438	2.5	6.0	52.92	1.0113 ± 0.0018	1.0149
TD356	2.865	3.2	4.5	33.07	0.8909 ± 0.0017	0.8943
TD353	2.865	3.2	5.0	36.75	0.9190 ± 0.0018	0.9226
TD357	2.865	3.2	5.5	40.42	0.9459 ± 0.0016	0.9491
TD354	2.865	3.2	6.0	44.10	0.9704 ± 0.0017	0.9738
TD358	2.865	3.2	6.5	47.77	0.9973 ± 0.0015	1.0003
TD362	2.292	4.2	5.0	29.40	0.8783 ± 0.0019	0.8821
TD366	2.292	4.2	5.5	32.34	0.9066 ± 0.0017	0.9100
TD363	2.292	4.2	6.0	35.28	0.9335 ± 0.0018	0.9371
TD367	2.292	4.2	6.5	38.22	0.9502 ± 0.0020	0.9542
TD364	2.292	4.2	7.0	41.16	0.9738 ± 0.0018	0.9774
TD368	2.292	4.2	7.5	44.10	0.9958 ± 0.0019	0.9996
TD369	2.292	4.2	8.0	47.04	1.0186 ± 0.0019	1.0224
TD376	1.948	5.1	5.5	27.48	0.8765 ± 0.0017	0.8799
TD373	1.948	5.1	6.0	29.98	0.8994 ± 0.0018	0.9030
TD377	1.948	5.1	6.5	32.48	0.9260 ± 0.0017	0.9294
TD374	1.948	5.1	7.0	34.98	0.9519 ± 0.0017	0.9553
TD378	1.948	5.1	7.5	37.48	0.9699 ± 0.0018	0.9735
TD375	1.948	5.1	8.0	39.98	0.9859 ± 0.0017	0.9893
TD379	1.948	5.1	8.5	42.48	1.0072 ± 0.0017	1.0106

Table 8. Infinite slab of water-saturated PuO₂, bounded above and below by water-saturated soil.

Case Name	Concentration (g PuO ₂ /cm ³)	H/X	Slab Thickness (cm)	$k_{\text{eff}} \pm 1\sigma$	$k_{\text{eff}} + 2\sigma$
TD003	4.584	3.9	1.0	0.8388 ± 0.0018	0.8424
TD007	4.584	3.9	1.5	0.9239 ± 0.0017	0.9273
TD004	4.584	3.9	2.0	0.9911 ± 0.0016	0.9943
TD005	4.584	3.9	2.5	1.0551 ± 0.0017	1.0585
TD013	4.000	4.9	1.0	0.8217 ± 0.0017	0.8251
TD017	4.000	4.9	1.5	0.9098 ± 0.0015	0.9128
TD014	4.000	4.9	2.0	0.9769 ± 0.0017	0.9803
TD015	4.000	4.9	2.5	1.0363 ± 0.0018	1.0399
TD023	3.438	6.1	1.0	0.8120 ± 0.0017	0.8154
TD027	3.438	6.1	1.5	0.8900 ± 0.0018	0.8936
TD024	3.438	6.1	2.0	0.9591 ± 0.0018	0.9627
TD025	3.438	6.1	2.5	1.0184 ± 0.0017	1.0218
TD037	2.865	7.9	1.5	0.8738 ± 0.0017	0.8772
TD034	2.865	7.9	2.0	0.9396 ± 0.0017	0.9430
TD035	2.865	7.9	2.5	0.9998 ± 0.0017	1.0032
TD047	2.292	10.5	1.5	0.8551 ± 0.0017	0.8585
TD044	2.292	10.5	2.0	0.9247 ± 0.0016	0.9279
TD045	2.292	10.5	2.5	0.9794 ± 0.0017	0.9828
TD046	2.292	10.5	3.0	1.0290 ± 0.0018	1.0326
TD057	1.948	12.8	1.5	0.8430 ± 0.0017	0.8464
TD054	1.948	12.8	2.0	0.9094 ± 0.0016	0.9126
TD055	1.948	12.8	2.5	0.9652 ± 0.0018	0.9688
TD059	1.948	12.8	2.82	0.9976 ± 0.0018	1.0012
TD056	1.948	12.8	3.0	1.0620 ± 0.0016	1.0178

Table 9. Infinite slab of PuO₂ and water-saturated soil, bounded above and below by water-saturated soil.

Case Name	Concentration (g PuO ₂ /cm ³)	H/X	Slab Thickness (cm)	$k_{\text{eff}} \pm 1\sigma$	$k_{\text{eff}} + 2\sigma$
TD263	4.584	1.6	1.5	0.8780 ± 0.0016	0.8812
TD262	4.584	1.6	2.0	0.9398 ± 0.0016	0.9430
TD264	4.584	1.6	2.5	0.9935 ± 0.0017	0.9969
TD261	4.584	1.6	3.0	1.0405 ± 0.0017	1.0439
TD273	4.000	2.0	1.5	0.8560 ± 0.0017	0.8594
TD272	4.000	2.0	2.0	0.9162 ± 0.0018	0.9198
TD274	4.000	2.0	2.5	0.9685 ± 0.0018	0.9721
TD271	4.000	2.0	3.0	1.0146 ± 0.0020	1.0186
TD285	3.438	2.5	2.0	0.8898 ± 0.0016	0.8930
TD283	3.438	2.5	2.5	0.9408 ± 0.0019	0.9446
TD281	3.438	2.5	3.0	0.9853 ± 0.0018	0.9889
TD284	3.438	2.5	3.5	1.0277 ± 0.0016	1.0309
TD293	2.865	3.2	2.0	0.8663 ± 0.0017	0.8695
TD294	2.865	3.2	2.5	0.9116 ± 0.0017	0.9148
TD291	2.865	3.2	3.0	0.9538 ± 0.0017	0.9572
TD295	2.865	3.2	3.5	0.9940 ± 0.0017	0.9974
TD292	2.865	3.2	4.0	1.0291 ± 0.0017	1.0321
TD303	2.292	4.2	2.5	0.8781 ± 0.0017	0.8815
TD301	2.292	4.2	3.0	0.9195 ± 0.0016	0.9227
TD304	2.292	4.2	3.5	0.9576 ± 0.0017	0.9610
TD302	2.292	4.2	4.0	0.9941 ± 0.0018	0.9977
TD305	2.292	4.2	4.5	1.0277 ± 0.0017	1.0311
TD313	1.948	5.1	2.5	0.8608 ± 0.0016	0.8640
TD311	1.948	5.1	3.0	0.9006 ± 0.0016	0.9038
TD314	1.948	5.1	3.5	0.9342 ± 0.0019	0.9380
TD312	1.948	5.1	4.0	0.9668 ± 0.0015	0.9698
TD317	1.948	5.1	4.42	0.9956 ± 0.0016	0.9988
TD315	1.948	5.1	4.5	1.0028 ± 0.0016	1.0060

Table 10. Results of water intrusion criticality scenarios.

Critical Configuration	Variables	Conclusion	Minimum critical PuO ₂
Water-moderated PuO ₂ sphere	Radius and PuO ₂ concentration	As concentration decreases critical mass decreases	8.4 kg in 7.96 in. diameter sphere
Water-saturated PuO ₂ and soil sphere	Radius and PuO ₂ concentration	As concentration decreases critical mass remains constant	22.6 kg in 8.31 in. diameter sphere
Slab of water-moderated PuO ₂ having the diameter of a 55-gallon drum (22.5 in.)	Slab thickness and the PuO ₂ concentration	As concentration decreased the critical mass decreased	23.2 kg in 4.6-cm-thick slab
Slab of water-saturated PuO ₂ and soil having the diameter of a 55-gallon drum (22.5 in.)	Slab thickness and the PuO ₂ concentration	As concentration decreased the critical mass decreased	41.0 kg in 8.3-cm-thick slab
Infinite slab of water-moderated PuO ₂	Slab thickness and the PuO ₂ concentration	As concentration decreased the critical thickness increased	2.8-cm-thick slab
Infinite slab of water-saturated PuO ₂ and soil	Slab thickness and the plutonium PuO ₂	As concentration decreased the critical thickness increased	4.4-cm-thick slab

6.6 Collection of Fissile Materials in the Off-Gas System

The last aspect of the proposed ISTD process that needs to be addressed from a criticality safety standpoint is the possibility of forming a critical configuration in the off-gas collection system.

Accumulation of sufficient fissile material in the off-gas system to cause a criticality event is not credible. The subsurface nature of ISTD is such that most of the fissile material remains in the soil. Sand between the heater and the slotted vacuum/heater well casing prevents particulate from entering the vacuum/heater well and serves as a roughing filter for the off-gas. The amount of plutonium that migrates from the treated soil, through the overburden and the sand filters will not be significant. Gram quantities of fissile material are not expected to enter the off-gas system. Only contamination levels of fissile material are expected. Any fissile material that does enter the system will be of insufficient quantity to cause a criticality. Therefore, a criticality accident is not credible in the off-gas system.

7. DESIGN FEATURES AND ADMINISTRATIVE CONTROLS AND LIMITS

There are no design features or administrative controls necessary in the application of the ISTD process.

8. SUMMARY AND CONCLUSIONS

Various configurations were evaluated to determine if any criticality concerns arise in conjunction with treating the buried waste contained in the SDA with the ISTD process. This evaluation consisted of three phases. The first phase was to consider criticality scenarios during the initial application of the ISTD process. The second phase consisted of evaluating the final configurations as a result of the application of the ISTD process. The third phase was to address ancillary issues relating to ISTD and criticality safety.

The first phase consisted of the evaluation of criticality concerns and postulated scenarios during the initial application of the ISTD process. They include the formation of a critical system resulting from the concentration of metal fissile material, and the combination of fissile material and melting polyethylene.

The formation of a critical system due to the concentration of metal fissile material was deemed to be not credible because of the low overall concentrations of fissile material within the waste, the high probability that oxidation of any metal has occurred, and the difficulty in reducing oxide to metal form due to ISTD.

Previous analysis (Sentieri 2003) demonstrates the fissile masses necessary to postulate a critical system composed of plutonium and polyethylene, in conjunction with the optimal geometry, reflection conditions, fissile concentration, and lack of diluent/absorber material. The amount of fissile mass necessary in a localized area, and the concurrent conditions necessary, lead to the conclusion that the formation of a critical system resulting from the initial application of the ISTD process is not credible.

The second phase consisted of evaluating the final expected waste configuration, due to the application of the ISTD process. This consisted of evaluating whether water reentry into the SDA after the ISTD process will cause the potential to create a critical system. Models were evaluated that consisted of fissile material dispersed in water and in water-saturated soil. These models show that unrealistically large fissile masses are necessary to form a critical system. The conclusion from these assessments was that none of these scenarios lend themselves to the credible formation of a critical system.

The collection of fissile material in the off-gas system was also evaluated. The conclusion from these assessments was that any fissile material that does enter the off-gas system would be of insufficient quantity to cause a criticality.

This evaluation concludes that there is no credible scenario associated with the ISTD process to formulate a critical system.

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Appendix A

Materials and Compositions

Appendix A

Materials and Compositions

Table A-1. INEEL soil sample analysis (average of two analyses).

Oxide	Composition (wt%)
SiO ₂	62.60
Al ₂ O ₃	11.85
Fe ₂ O ₃	4.25
CaO	3.68
K ₂ O	2.99
MgO	1.72
Na ₂ O	1.37
TiO ₂	0.68
MnO ₂	0.10
BaO	0.09
ZrO ₂	0.05
B ₂ O ₃	0.05
NiO	0.04
SrO	0.02
Cr ₂ O ₃	0.02
Total Oxide	89.5%
Moisture	7.5%

Table A-2. Renormalized INEEL Soil Sample for Dry Material.

Oxide	Composition (wt%)
SiO ₂	69.936
Al ₂ O ₃	13.239
Fe ₂ O ₃	4.748
CaO	4.111
K ₂ O	3.340
MgO	1.922
Na ₂ O	1.531
TiO ₂	0.760
MnO ₂	0.112
BaO	0.101
ZrO ₂	0.056
B ₂ O ₃	0.056
NiO	0.044
SrO	0.022
Cr ₂ O ₃	0.022
Total Oxide	100.0%

Table A-3. Material compositions of water saturated PuO₂.

Concentration (g PuO ₂ /cm ³)	Added water H/X	Element	Atom Density (atoms/b-cm)
4.584	3.9	Pu-239	1.01845x10 ⁻²
		H	4.00411x10 ⁻²
		O	4.03896x10 ⁻²
4.000	4.9	Pu-239	8.88602x10 ⁻³
		H	4.34446x10 ⁻²
		O	3.94943x10 ⁻²
3.438	6.1	Pu-239	7.63841x10 ⁻³
		H	4.67146x10 ⁻²
		O	3.86341x10 ⁻²

Concentration (g PuO ₂ /cm ³)	Added water H/X	Element	Atom Density (atoms/b-cm)
2.865	7.9	Pu-239	6.36534x10 ⁻³
		H	5.00513x10 ⁻²
		O	3.77564x10 ⁻²
2.292	10.5	Pu-239	5.09227x10 ⁻³
		H	5.33881x10 ⁻²
		O	3.68786x10 ⁻²
1.948	12.8	Pu-239	4.32843x10 ⁻³
		H	5.53902x10 ⁻²
		O	3.63519x10 ⁻²

Table A-4. Material compositions of PuO₂ and water saturated soil

Concentration (g PuO ₂ /cm ³)	Added water H/X	Element	Atom Density (atoms/b-cm)
4.584	1.6	B-11	8.2674x10 ⁻⁶
		Na	2.5555x10 ⁻⁴
		Mg	2.4661x10 ⁻⁴
		Al	1.3432x10 ⁻³
		Si	6.0204x10 ⁻³
		K	3.6678x10 ⁻⁴
		Ca	3.7918x10 ⁻⁴
		Ti	4.9202x10 ⁻⁵
		Mn	6.6648x10 ⁻⁶
		Fe	3.0758x10 ⁻⁴
		Pu-239	1.01845x10 ⁻²
		H	1.6045x10 ⁻²
		O	4.3970x10 ⁻²
4.000	2.0	B-11	8.9701x10 ⁻⁶
		Na	2.7727x10 ⁻⁴
		Mg	2.6757x10 ⁻⁴
		Al	1.4574x10 ⁻³
		Si	6.5321x10 ⁻³
		K	3.9796x10 ⁻⁴
		Ca	4.1141x10 ⁻⁴
		Ti	5.3385x10 ⁻⁵
		Mn	7.2313x10 ⁻⁶
		Fe	3.3372x10 ⁻⁴
		Pu-239	8.88602x10 ⁻³
		H	1.7409x10 ⁻²
		O	4.3379x10 ⁻²

Concentration (g PuO ₂ /cm ³)	Added water H/X	Element	Atom Density (atoms/b-cm)
3.438	2.5	B-11	9.6453x10 ⁻⁶
		Na	2.9814x10 ⁻⁴
		Mg	2.8771x10 ⁻⁴
		Al	1.5671x10 ⁻³
		Si	7.0238x10 ⁻³
		K	4.2791x10 ⁻⁴
		Ca	4.4237x10 ⁻⁴
		Ti	5.7403x10 ⁻⁵
		Mn	7.7756x10 ⁻⁶
		Fe	3.5884x10 ⁻⁴
		Pu-239	7.63841x10 ⁻³
		H	1.8719x10 ⁻²
		O	4.2811x10 ⁻²
2.865	3.2	B-11	1.0334x10 ⁻⁵
		Na	3.1943x10 ⁻⁴
		Mg	3.0827x10 ⁻⁴
		Al	1.6790x10 ⁻³
		Si	7.5255x10 ⁻³
		K	4.5848x10 ⁻⁴
		Ca	4.7397x10 ⁻⁴
		Ti	6.1503x10 ⁻⁵
		Mn	8.3310x10 ⁻⁶
		Fe	3.8447x10 ⁻⁴
		Pu-239	6.36534x10 ⁻³
		H	2.0057x10 ⁻²
		O	4.2232x10 ⁻²
2.292	4.2	B-11	1.1023x10 ⁻⁵
		Na	3.4073x10 ⁻⁴
		Mg	3.2882x10 ⁻⁴
		Al	1.7910x10 ⁻³
		Si	8.0272x10 ⁻³
		K	4.8904x10 ⁻⁴
		Ca	5.0557x10 ⁻⁴
		Ti	6.5603x10 ⁻⁵
		Mn	8.8864x10 ⁻⁶
		Fe	4.1010x10 ⁻⁴
		Pu-239	5.09227x10 ⁻³
		H	2.1394x10 ⁻²
		O	4.1653x10 ⁻²

Concentration (g PuO ₂ /cm ³)	Added water H/X	Element	Atom Density (atoms/b-cm)
1.948	5.1	B-11	1.1437x10 ⁻⁵
		Na	3.5351x10 ⁻⁴
		Mg	3.4115x10 ⁻⁴
		Al	1.8581x10 ⁻³
		Si	8.3282x10 ⁻³
		K	5.0738x10 ⁻⁴
		Ca	5.2453x10 ⁻⁴
		Ti	6.8063x10 ⁻⁵
		Mn	9.2196x10 ⁻⁶
		Fe	4.2548x10 ⁻⁴
		Pu-239	4.32843x10 ⁻³
		H	2.2196x10 ⁻²
		O	4.1305x10 ⁻²

Appendix B

Supporting Calculations and Methods

Appendix B

Supporting Calculations and Methods

B-1. CONTAMINATED SOIL CALCULATIONS

Consider a sample of soil with volume V_{slag} and density ρ_{slag} . It has mass m_{slag} and is composed entirely of oxides. We know the mass fraction f_i of each oxide in the soil, so we know the mass of each oxide in the sample. Consider SiO_2 .

$$m_{SiO_2} = f_{SiO_2} m_{slag}, \quad M_{SiO_2} = M_{Si} + 2M_O \quad \text{and} \quad m_{slag} = \rho_{slag} V_{slag} \quad \text{so that}$$

$$m_{Si} = \left(\frac{M_{Si}}{M_{SiO_2}} \right) m_{SiO_2} = f_{SiO_2} \left(\frac{M_{Si}}{M_{SiO_2}} \right) \rho_{slag} V_{slag}.$$

Now consider adding PuO_2 with density ρ_{PuO_2} to the soil. Let $\mu \equiv \frac{m_{PuO_2}}{V_{mix}}$ be the concentration (g/cc) of PuO_2 in the mix. Now $V_{mix} = V_{slag} + V_{PuO_2}$ and $m_{PuO_2} = \mu V_{mix}$.

$$m_{mix} = m_{slag} + m_{PuO_2} = m_{slag} + \mu V_{mix} = \rho_{slag} V_{slag} + \mu V_{mix}.$$

$$\rho_{mix} = \frac{m_{mix}}{V_{mix}} = \frac{\rho_{slag} V_{slag}}{V_{mix}} + \mu, \quad \rho_{PuO_2} V_{PuO_2} = m_{PuO_2} = \mu V_{mix} \quad \text{and} \quad V_{mix} = \frac{m_{PuO_2}}{\mu}.$$

$$V_{mix} = \frac{m_{PuO_2}}{\mu} = V_{slag} + V_{PuO_2} = \frac{\rho_{PuO_2} V_{PuO_2}}{\mu} \quad \text{so that} \quad V_{PuO_2} \left[\frac{\rho_{PuO_2}}{\mu} - 1 \right] = V_{slag}. \quad (1)$$

Combining (1) with $V_{mix} = V_{slag} + V_{PuO_2}$ gives

$$V_{mix} = V_{slag} \left[\frac{\rho_{PuO_2}}{\rho_{PuO_2} - \mu} \right].$$

Then

$$\rho_{mix} = \frac{\rho_{slag} V_{slag}}{V_{mix}} + \mu \quad \text{and} \quad \frac{V_{slag}}{V_{mix}} = \left(\frac{\rho_{PuO_2} - \mu}{\rho_{PuO_2}} \right).$$

so that

$$\rho_{mix} = \rho_{slag} \left(\frac{\rho_{PuO_2} - \mu}{\rho_{PuO_2}} \right) + \mu.$$

$$V_{PuO_2} = \left(\frac{\mu}{\rho_{PuO_2} - \mu} \right) V_{slag}.$$

From (1) we have

Let $\xi \equiv \frac{m_{Pu}}{V_{mix}}$ be the concentration (g/cc) of Pu in the mix. We have $m_{Pu} = \left(\frac{M_{Pu}}{M_{PuO_2}} \right) m_{PuO_2}$ so

that $\xi = \left(\frac{M_{Pu}}{M_{PuO_2}} \right) \frac{m_{PuO_2}}{V_{mix}} = \left(\frac{M_{Pu}}{M_{PuO_2}} \right) \mu.$

We thus have

$$\mu = \left(\frac{M_{PuO_2}}{M_{Pu}} \right) \xi$$

Now $m_{Si} = f_{SiO_2} \left(\frac{M_{Si}}{M_{SiO_2}} \right) \rho_{slag} V_{slag}$ and $m_{mix} = \rho_{mix} V_{mix} = \rho_{mix} V_{slag} \left(\frac{\rho_{PuO_2}}{\rho_{PuO_2} - \mu} \right)$ so that

$$\frac{m_{Si}}{m_{mix}} = \frac{f_{SiO_2} \left(\frac{M_{Si}}{M_{SiO_2}} \right) \rho_{slag} V_{slag}}{\rho_{mix} V_{slag} \left(\frac{\rho_{PuO_2}}{\rho_{PuO_2} - \mu} \right)} = f_{SiO_2} \left(\frac{M_{Si}}{M_{SiO_2}} \right) \left(\frac{\rho_{slag}}{\rho_{mix}} \right) \left(\frac{\rho_{PuO_2} - \mu}{\rho_{PuO_2}} \right)$$

and the weight fraction of Silicon in the mix is

$$F_{Si} = f_{SiO_2} \left(\frac{M_{Si}}{M_{SiO_2}} \right) \left(\frac{\rho_{slag}}{\rho_{mix}} \right) \left(\frac{\rho_{PuO_2} - \mu}{\rho_{PuO_2}} \right)$$

$$F_{Pu} = \frac{m_{Pu}}{m_{mix}} = \left(\frac{M_{Pu}}{M_{PuO_2}} \right) \frac{m_{PuO_2}}{m_{mix}} = \left(\frac{M_{Pu}}{M_{PuO_2}} \right) \frac{\mu}{\rho_{mix}} = \left(\frac{M_{Pu}}{M_{PuO_2}} \right) \left(\frac{M_{PuO_2}}{M_{Pu}} \right) \frac{\xi}{\rho_{mix}} = \frac{\xi}{\rho_{mix}}.$$

Now consider the mass fraction of oxygen contributed by the PuO_2 .

$$m_{\text{PuO}_2} = m_{\text{Pu}} + m_{\text{O}}^{\text{PuO}_2} \quad \text{so that} \quad \frac{m_{\text{PuO}_2}}{m_{\text{mix}}} = \frac{m_{\text{Pu}}}{m_{\text{mix}}} + \frac{m_{\text{O}}^{\text{PuO}_2}}{m_{\text{mix}}} = F_{\text{Pu}} + F_{\text{O}}^{\text{PuO}_2}.$$

$$\text{Now } \mu = \frac{m_{\text{PuO}_2}}{V_{\text{mix}}} \quad \text{so that} \quad \frac{m_{\text{PuO}_2}}{m_{\text{mix}}} = \frac{\mu V_{\text{mix}}}{m_{\text{mix}}} = \frac{\mu}{\rho_{\text{mix}}}, \quad \frac{\mu}{\rho_{\text{mix}}} = F_{\text{Pu}} + F_{\text{O}}^{\text{PuO}_2} \quad \text{and} \quad F_{\text{O}}^{\text{PuO}_2} = \frac{\mu}{\rho_{\text{mix}}} - F_{\text{Pu}}$$

$$\text{but } \mu = \left(\frac{M_{\text{PuO}_2}}{M_{\text{Pu}}} \right) \xi \quad \text{so that we have} \quad F_{\text{O}}^{\text{PuO}_2} = \frac{1}{\rho_{\text{mix}}} \left(\frac{M_{\text{PuO}_2}}{M_{\text{Pu}}} \right) \xi - F_{\text{Pu}}$$

Consider the oxygen contribution from SiO_2 . We have 2 oxygen molecules/molecule so that the mass fraction in the mix is given by

$$F_{\text{O}}^{\text{SiO}_2} = f_{\text{SiO}_2} \left(\frac{2M_{\text{O}}}{M_{\text{SiO}_2}} \right) \left(\frac{\rho_{\text{slag}}}{\rho_{\text{mix}}} \right) \left(\frac{\rho_{\text{PuO}_2} - \mu}{\rho_{\text{PuO}_2}} \right) = f_{\text{SiO}_2} \left(\frac{2M_{\text{O}}}{M_{\text{SiO}_2}} \right) \varphi.$$

The oxygen contributions from the other oxides are calculated similarly.

Now consider atomic fractions and total atom density. We have

$$N_{\text{Si}} = \frac{m_{\text{Si}} (\text{g})}{M_{\text{Si}} (\text{g/mole})} N_A (\text{@/mole}) \quad n_{\text{Si}} = \frac{N_{\text{Si}}}{V_{\text{mix}}} = \frac{m_{\text{Si}}}{V_{\text{mix}}} \frac{N_A}{M_{\text{Si}}}$$

$$\text{but } m_{\text{Si}} = F_{\text{Si}} m_{\text{mix}} \quad \text{so that} \quad \frac{m_{\text{Si}}}{V_{\text{mix}}} = F_{\text{Si}} \frac{m_{\text{mix}}}{V_{\text{mix}}} = F_{\text{Si}} \rho_{\text{mix}} \quad \text{and thus, in general,}$$

$$n_{\text{Si}} = F_{\text{Si}} \rho_{\text{mix}} \frac{N_A}{M_{\text{Si}}}$$

$$n_{\text{cation}_i} = F_{\text{cation}_i} \rho_{\text{mix}} \left(\frac{N_A}{M_{\text{cation}_i}} \right) \quad \text{and} \quad n_{\text{Pu}} = F_{\text{Pu}} \rho_{\text{mix}} \left(\frac{N_A}{M_{\text{Pu}}} \right) \quad n_{\text{O}} = \sum_i n_{\text{O}_i}$$

$$n_{\text{mix}} = \sum_i n_{\text{cation}_i} + n_{\text{Pu}} + \sum_i n_{\text{O}_i}$$

Let β_i be the atomic fraction for cation_i .

$$\beta_{\text{cation}_i} = \frac{n_{\text{cation}_i}}{n_{\text{mix}}} \quad \beta_{\text{Pu}} = \frac{n_{\text{Pu}}}{n_{\text{mix}}} \quad \beta_{\text{O}} = \frac{n_{\text{O}}}{n_{\text{mix}}}.$$

Now $m_{SiO_2}^O \equiv$ mass of oxygen from SiO_2 , and

$$m_{SiO_2}^O = \left(\frac{2M_O}{M_{SiO_2}} \right) m_{SiO_2} = \left(\frac{2M_O}{M_{SiO_2}} \right) f_{SiO_2} m_{slag} = \left(\frac{2M_O}{M_{SiO_2}} \right) f_{SiO_2} \rho_{slag} V_{slag}$$

$$\frac{m_{SiO_2}^O}{m_{mix}} = \frac{m_{SiO_2}^O}{\rho_{mix} V_{mix}} = \left(\frac{2M_O}{M_{SiO_2}} \right) f_{SiO_2} \frac{\rho_{slag} V_{slag}}{\rho_{mix} V_{mix}} \quad \text{but} \quad \frac{V_{slag}}{V_{mix}} = \left(\frac{\rho_{PuO_2} - \mu}{\rho_{PuO_2}} \right) \quad \text{so that}$$

$$\frac{m_{SiO_2}^O}{m_{mix}} = \left(\frac{2M_O}{M_{SiO_2}} \right) f_{SiO_2} \frac{\rho_{slag}}{\rho_{mix}} \left(\frac{\rho_{PuO_2} - \mu}{\rho_{PuO_2}} \right).$$

Calculate the corresponding quantity for the other oxides and PuO_2 and add them up to get F_O .

Procedure

1. Specify ξ
2. Calculate μ .
3. For given ρ_{slag} , ρ_{PuO_2} , calculate ρ_{mix} .
4. Calculate
$$\phi \equiv \left(\frac{\rho_{slag}}{\rho_{mix}} \right) \left(\frac{\rho_{PuO_2} - \mu}{\rho_{PuO_2}} \right)$$
5. Given the mass fractions of the oxides in the soil, calculate the mass fractions F_{cation_i} of the cations in the mix.
6. Calculate the mass fraction for the plutonium: F_{Pu} .
7. Calculate the atomic densities n_{cation_i} from the mass fractions.
8. Calculate the total atom density n_O for oxygen by summing the atomic densities n_i^O from each oxide.
9. Calculate the total atom density for the mixture by summing the atom densities of the constituents.
10. Calculate the atomic fractions β_{cation_i} , β_{Pu} and β_O .

B-2. SATURATED POROUS SOIL CALCULATIONS

Consider V_s of solids with mass $m_s = \rho_s V_s$ and with 40% porosity. We have $V_s = 0.6V_T$ where V_T is the total volume occupied by the porous soil. We then have

$$V_T = \frac{V_s}{0.6} = \frac{5}{3}V_s$$

The water filling the pores occupies the rest:

$$V_w = 0.4V_T = 0.4\left(\frac{V_s}{0.6}\right) = \frac{2}{3}V_s$$

$$m_T = m_s + m_w = \rho_s V_s + \rho_w V_w \text{ but } \rho_w = 1.$$

$$m_T = \rho_s V_s + \frac{2}{3}V_s = V_s\left(\rho_s + \frac{2}{3}\right) \text{ and } \rho_{soil} = \frac{m_T}{V_T} = \frac{V_s\left(\rho_s + \frac{2}{3}\right)}{\frac{5}{3}V_s} = \frac{3}{5}\left(\rho_s + \frac{2}{3}\right) = \left(\frac{3\rho_s + 2}{5}\right).$$

The mass fraction of SiO_2 in the solids is $f_{\text{SiO}_2} = \frac{m_{\text{SiO}_2}}{m_s}$ so that $m_{\text{SiO}_2} = f_{\text{SiO}_2} m_s = f_{\text{SiO}_2} \rho_s V_s$ thus

$$m_{\text{Si}} = \left(\frac{M_{\text{Si}}}{M_{\text{SiO}_2}}\right) m_{\text{SiO}_2} = f_{\text{SiO}_2} \left(\frac{M_{\text{Si}}}{M_{\text{SiO}_2}}\right) \rho_s V_s$$

$$F_{\text{Si}} = f_{\text{SiO}_2} \left(\frac{M_{\text{Si}}}{M_{\text{SiO}_2}}\right) \frac{\rho_s V_s}{V_s\left(\rho_s + \frac{2}{3}\right)} = f_{\text{SiO}_2} \left(\frac{M_{\text{Si}}}{M_{\text{SiO}_2}}\right) \left(\frac{\rho_s}{\rho_s + \frac{2}{3}}\right)$$

$$F_{\text{Si}} = f_{\text{SiO}_2} \left(\frac{M_{\text{Si}}}{M_{\text{SiO}_2}}\right) \left(\frac{3\rho_s}{3\rho_s + 2}\right).$$

$$\text{Now } \frac{m_o^{\text{SiO}_2}}{m_T} = F_o^{\text{SiO}_2} \text{ but } m_o^{\text{SiO}_2} = \left(\frac{2M_o}{M_{\text{SiO}_2}}\right) m_{\text{SiO}_2} \text{ and } m_o^{\text{SiO}_2} = f_{\text{SiO}_2} \left(\frac{2M_o}{M_{\text{SiO}_2}}\right) \rho_s V_s$$

$$F_o^{\text{SiO}_2} = f_{\text{SiO}_2} \left(\frac{2M_o}{M_{\text{SiO}_2}}\right) \frac{\rho_s V_s}{V_s\left(\rho_s + \frac{2}{3}\right)} \text{ or } F_o^{\text{SiO}_2} = f_{\text{SiO}_2} \left(\frac{2M_o}{M_{\text{SiO}_2}}\right) \left(\frac{3\rho_s}{3\rho_s + 2}\right).$$

Now
$$F_o^{H_2O} = \frac{m_o^{H_2O}}{m_T} = \frac{1}{m_T} \left(\frac{M_o}{M_{H_2O}} \right) m_w \quad \text{but } m_w = \rho_w V_w \text{ and } \rho_w = 1 \text{ so that}$$

$$F_o^{H_2O} = \frac{1}{V_s \left(\rho_s + \frac{2}{3} \right)} \left(\frac{M_o}{M_{H_2O}} \right) \frac{2}{3} V_s = \left(\frac{M_o}{M_{H_2O}} \right) \left(\frac{2}{3\rho_s + 2} \right)$$

$$F_H = \left(\frac{2M_H}{M_{H_2O}} \right) \left(\frac{2}{3\rho_s + 2} \right)$$

$$n_H \text{ (@ / B } \times \text{ cm)} = \frac{F_H \rho_s}{M_H} N_A \times 10^{-24} .$$

B-3. CONTAMINATED SATURATED SOIL CALCULATIONS

CSS	=	Contaminated Saturated Soil
V_s	=	Volume of solids in the porous soil
V_{ss}	=	Volume of saturated soil
V_w	=	Volume of water in the pores
V_{css}	=	Volume of contaminated saturated soil
ρ_s	=	Mass density of compacted soil
ρ_{ss}	=	Mass density of clean saturated soil
ρ_{css}	=	Mass density of contaminated saturated soil
ρ_w	=	Mass density of water (1.0 g/cc)
ρ_{PuO_2}	=	Theoretical mass density of PuO_2 (11.46 g/cc)
m_{ss}	=	Mass of saturated soil
m_w	=	Mass of water in soil
m_s	=	Mass of soil solids
m_{css}	=	Mass of contaminated saturated soil

m_{Si} = Mass of Silicon present

m_{SiO_2} = Mass of SiO_2 present

$m_O^{H_2O}$ = Mass of oxygen contributed by H_2O

M_i = Molecular weight (g/mole) of substance i

F_{Si} = Mass fraction of Si in CSS

$F_O^{SiO_2}$ = Mass fraction of oxygen contributed by SiO_2

f_{SiO_2} = Mass fraction is SiO_2 in the dry soil

μ = Concentration of PuO_2 in CSS

ξ = Concentration of Pu in CSS

N_A = Avogadroes number

$m_{ss} = \rho_s V_s + \rho_w V_w$ but $\rho_w = 1$, and $V_{ss} = V_s + V_w$, and for 40% porosity we have

$V_s = 0.6V_{ss}$, $V_w = 0.4V_{ss}$ thus $V_{ss} = \frac{V_s}{0.6} = \frac{5}{3}V_s$. $V_w = 0.4V_{ss} = \frac{2}{3}V_s$ so that

$$m_{ss} = \rho_s V_s + \frac{2}{3}V_s = V_s \left(\rho_s + \frac{2}{3} \right).$$

Now $\xi \equiv \frac{m_{Pu}}{V_{CSS}}$ and $\mu \equiv \frac{m_{PuO_2}}{V_{CSS}}$ and thus $\mu = \left(\frac{M_{PuO_2}}{M_{Pu}} \right) \xi$

$$m_{CSS} = m_{ss} + m_{PuO_2} = m_{ss} + \mu V_{CSS} \quad \text{and} \quad \rho_{CSS} \equiv \frac{m_{CSS}}{V_{CSS}} = \frac{m_{ss}}{V_{CSS}} + \mu.$$

Substitution then gives

$$\rho_{ss} = \left(\frac{3\rho_s + 2}{5} \right)$$

Now $V_{CSS} = V_{SS} + V_{PuO_2}$, $V_{PuO_2} = \frac{m_{PuO_2}}{\rho_{PuO_2}}$ and $V_{SS} = \frac{5}{3}V_s$. $m_{PuO_2} = \mu V_{CSS}$ thus

$V_{CSS} = \frac{5}{3}V_s + \frac{\mu V_{CSS}}{\rho_{PuO_2}}$ which can be solved for V_{CSS} and manipulated to give

$$\frac{V_s}{V_{CSS}} = \frac{3}{5} \left(1 - \frac{\mu}{\rho_{PuO_2}} \right)$$

$$\rho_{CSS} = \frac{m_{SS}}{V_{CSS}} + \mu = \frac{V_s}{V_{CSS}} \left(\rho_s + \frac{2}{3} \right) + \mu$$

or

$$\rho_{CSS} = \mu + \frac{3}{5} \left(1 - \frac{\mu}{\rho_{PuO_2}} \right) \left(\rho_s + \frac{2}{3} \right)$$

which reduces to

$$\rho_{CSS} = \mu + \left(1 - \frac{\mu}{\rho_{PuO_2}} \right) \rho_{SS}.$$

Now

$$m_{Si} = \left(\frac{M_{Si}}{M_{SiO_2}} \right) m_{SiO_2} = f_{SiO_2} \left(\frac{M_{Si}}{M_{SiO_2}} \right) \rho_s V_s \quad F_{Si} = \frac{m_{Si}}{m_{CSS}}, \quad \rho_{CSS} V_{CSS} = m_{CSS}$$

so that

$$F_{Si} = \frac{\left(\frac{M_{Si}}{M_{SiO_2}} \right) f_{SiO_2} \rho_s V_s}{\rho_{CSS} V_{CSS}} = \left(\frac{V_s}{V_{CSS}} \right) \frac{\rho_s}{\rho_{CSS}} \left(\frac{M_{Si}}{M_{SiO_2}} \right) f_{SiO_2} \quad \text{thus}$$

$$F_{Si} = \frac{3}{5} \left(1 - \frac{\mu}{\rho_{PuO_2}} \right) \left(\frac{\rho_s}{\rho_{CSS}} \right) \left(\frac{M_{Si}}{M_{SiO_2}} \right) f_{SiO_2}$$

or

$$F_{Si} = \frac{3}{5} \left(\frac{M_{Si}}{M_{SiO_2}} \right) \left(\frac{\rho_s}{\rho_{CSS}} \right) f_{SiO_2} \left(1 - \frac{\mu}{\rho_{PuO_2}} \right).$$

Now

$$F_o^{SiO_2} = \frac{3}{5} \left(\frac{2M_o}{M_{SiO_2}} \right) \left(\frac{\rho_s}{\rho_{CSS}} \right) f_{SiO_2} \left(1 - \frac{\mu}{\rho_{PuO_2}} \right).$$

Now consider the water.

$$F_o^{H_2O} \equiv \frac{m_o^{H_2O}}{m_{CSS}} = \frac{1}{m_{CSS}} \left(\frac{M_o}{M_{H_2O}} \right) m_w$$

but $m_w = \rho_w V_w$ and $\rho_w = 1$ so that $m_w = V_w = \frac{2}{3} V_s$ and $m_{CSS} = \rho_{CSS} V_{CSS}$

$$F_o^{H_2O} = \left(\frac{M_o}{M_{H_2O}} \right) \frac{2}{3} V_s \frac{1}{\rho_{CSS} V_{CSS}} = \left(\frac{V_s}{V_{CSS}} \right) \frac{2}{3 \rho_{CSS}} \left(\frac{M_o}{M_{H_2O}} \right)$$

then

$$F_o^{H_2O} = \frac{2}{3 \rho_{CSS}} \left(\frac{M_o}{M_{H_2O}} \right) \frac{3}{5} \left(1 - \frac{\mu}{\rho_{PuO_2}} \right)$$

or finally,

$$F_o^{H_2O} = \frac{2}{5 \rho_{CSS}} \left(\frac{M_o}{M_{H_2O}} \right) \left(1 - \frac{\mu}{\rho_{PuO_2}} \right)$$

$$F_H^{H_2O} = \frac{2}{5 \rho_{CSS}} \left(\frac{2M_H}{M_{H_2O}} \right) \left(1 - \frac{\mu}{\rho_{PuO_2}} \right).$$

There are no other hydrogen contributors to the mixture, so we have $F_H = F_H^{H_2O}$.

$$F_H = \frac{2}{5 \rho_{CSS}} \left(\frac{2M_H}{M_{H_2O}} \right) \left(1 - \frac{\mu}{\rho_{PuO_2}} \right)$$

The density of hydrogen in the mixture is $F_H \rho_{CSS}$ thus

$$n_H (@/B \times cm) = \frac{F_H \rho_{CSS} N_A}{M_H} \times 10^{-24}.$$

The contribution of the other oxides to the oxygen are calculated and summed to obtain the total oxygen mass fraction. The contribution of the other cations are calculated similarly to the case of silicon. Consider the plutonium oxide.

$$F_{Pu} \equiv \frac{m_{Pu}}{m_{CSS}} = \frac{1}{m_{CSS}} \left(\frac{M_{Pu}}{M_{PuO_2}} \right) m_{PuO_2} \quad \text{but } m_{CSS} = \rho_{CSS} V_{CSS} \text{ and } m_{PuO_2} = \mu V_{CSS} \text{ so that}$$

substituting gives

$$F_{Pu} = \frac{1}{\rho_{CSS} V_{CSS}} \left(\frac{M_{Pu}}{M_{PuO_2}} \right) \mu V_{CSS} = \frac{\mu}{\rho_{CSS}} \left(\frac{M_{Pu}}{M_{PuO_2}} \right)$$

and thus

$$F_{Pu} = \frac{\mu}{\rho_{CSS}} \left(\frac{M_{Pu}}{M_{PuO_2}} \right)$$

$$F_O^{PuO_2} \equiv \frac{m_O^{PuO_2}}{m_{CSS}} = \frac{1}{m_{CSS}} \left(\frac{2M_O}{M_{PuO_2}} \right) m_{PuO_2} = \frac{1}{\rho_{CSS} V_{CSS}} \left(\frac{2M_O}{M_{PuO_2}} \right) \mu V_{CSS}$$

so that

$$F_O^{PuO_2} = \left(\frac{\mu}{\rho_{CSS}} \right) \left(\frac{2M_O}{M_{PuO_2}} \right)$$

$$n_{Pu} (@/B \times cm) = \frac{F_{Pu} \rho_{CSS} N_A}{M_{Pu}} \times 10^{-24}.$$

Substituting $\mu = 0$ in the above expressions causes them to reduce to the simple forms of the derivation for clean 40% porous saturated soil. For $\mu \rightarrow 0$ we have

$$\frac{V_S}{V_{CSS}} \rightarrow \frac{3}{5}, \quad \frac{\rho_S}{\rho_{CSS}} \rightarrow \frac{5\rho_S}{3\rho_S + 2}, \quad F_{Si} \rightarrow \frac{3}{5} \left(\frac{M_{Si}}{M_{SiO_2}} \right) \left(\frac{\rho_S}{\rho_{CSS}} \right) f_{SiO_2}. \text{ Substitution gives}$$

$$F_{Si} \rightarrow \frac{3}{5} \left(\frac{M_{Si}}{M_{SiO_2}} \right) f_{SiO_2} \frac{5\rho_s}{3\rho_s + 2} = f_{SiO_2} \left(\frac{M_{Si}}{M_{SiO_2}} \right) \left(\frac{3\rho_s}{3\rho_s + 2} \right)$$

which agrees with the previous derivation.

$$F_o^{SiO_2} \rightarrow f_{SiO_2} \left(\frac{2M_o}{M_{SiO_2}} \right) \frac{3}{5} \left(\frac{5\rho_s}{3\rho_s + 2} \right) = f_{SiO_2} \left(\frac{2M_o}{M_{SiO_2}} \right) \left(\frac{3\rho_s}{3\rho_s + 2} \right)$$

$$F_o^{H_2O} \rightarrow \frac{2}{5\rho_{CSS}} \left(\frac{M_o}{M_{H_2O}} \right) = \frac{2}{5} \left(\frac{M_o}{M_{H_2O}} \right) \frac{5}{3\rho_s + 2} = \left(\frac{M_o}{M_{H_2O}} \right) \left(\frac{2}{3\rho_s + 2} \right)$$

$$F_H \rightarrow \frac{2}{5\rho_{CSS}} \left(\frac{2M_H}{M_{H_2O}} \right) = \frac{2}{5} \left(\frac{2M_H}{M_{H_2O}} \right) \frac{5}{3\rho_s + 2} = \left(\frac{2M_H}{M_{H_2O}} \right) \left(\frac{2}{3\rho_s + 2} \right)$$

It can be seen that these expressions all agree with the previous derivation.

B-4. WET PUO₂ CALCULATIONS

ρ_{pwr} = Packing density of PuO₂ (4.0 g/cc)

ρ_{PuO_2} = Theoretical PuO₂ density (11.46 g/cc)

ρ_{mix} = Saturated PuO₂ density

$m_O^{PuO_2}$ = Mass of oxygen contributed by PuO₂

$m_O^{H_2O}$ = Mass of oxygen contributed by H₂O

M_i = Molecular weight of substance i

m_i = Mass of substance i

V_i = Volume of substance i

F_o = Mass fraction of oxygen in the mix

F_H = Mass fraction of hydrogen in the mix

F_{Pu} = Mass fraction of plutonium in the mix

F_{PuO_2} = Mass fraction of PuO₂ in the mix

F_{H_2O} = Mass fraction of H_2O in the mix

n_O = Atomic density of oxygen in the mix (@/B × cm)

n_H = Atomic density of hydrogen in the mix (@/B × cm)

n_{Pu} = Atomic density of plutonium in the mix (@/B × cm)

n_T = Total atom density in the mix ($n_O + n_H + n_{Pu}$)

N_A = Avogadroes number

$$\rho_{pwr} = \frac{m_{PuO_2}}{V_{PuO_2}} \quad \text{or} \quad V_{PuO_2} = \frac{m_{PuO_2}}{\rho_{PuO_2}}, \quad m_{PuO_2} = \rho_{PuO_2} V_{PuO_2} \quad \text{and} \quad \rho_{pwr} = \rho_{PuO_2} \left(\frac{V_{PuO_2}}{V_{pwr}} \right)$$

$$\frac{V_{PuO_2}}{V_{pwr}} = \frac{\rho_{pwr}}{\rho_{PuO_2}} = \frac{4.0}{11.46} = 0.3490 \quad \text{and} \quad V_{pwr} = V_{mix}$$

The solid is only 34.9% of the volume. The rest is pore space, to be occupied by water.

$$m_{mix} = \rho_{PuO_2} V_{PuO_2} + \rho_{H_2O} V_{H_2O} \quad \text{but} \quad \rho_{H_2O} = 1.0$$

and

$$V_{PuO_2} = 0.3490 V_{pwr} = 0.3490 V_{mix}, \quad V_{H_2O} = 0.6510 V_{pwr} = 0.6510 V_{mix}$$

so that

$$m_{mix} = \rho_{PuO_2} (0.349 V_{mix}) + 0.651 V_{mix} \quad m_{mix} = (0.349 \rho_{PuO_2} + 0.651) V_{mix} = 4.6505 V_{mix}$$

$$\rho_{mix} = \frac{m_{mix}}{V_{mix}} = 4.6505 \text{ g/cc.}$$

Now consider the mass fractions.

$$m_{mix} = m_{PuO_2} + m_{H_2O} \quad m_{Pu} = \left(\frac{M_{Pu}}{M_{PuO_2}} \right) m_{PuO_2} \quad m_O^{PuO_2} = \left(\frac{2M_O}{M_{PuO_2}} \right) m_{PuO_2}$$

$$m_H = \left(\frac{2M_H}{M_{H_2O}} \right) m_{H_2O} \quad m_O^{H_2O} = \left(\frac{M_O}{M_{H_2O}} \right) m_{H_2O} \quad m_O = m_O^{H_2O} + m_O^{PuO_2}$$

$$F_{Pu} = \frac{m_{Pu}}{m_{mix}} = \left(\frac{M_{Pu}}{M_{PuO_2}} \right) \frac{m_{PuO_2}}{m_{mix}} = \left(\frac{M_{Pu}}{M_{PuO_2}} \right) F_{PuO_2}$$

$$F_H = \left(\frac{2M_H}{M_{H_2O}} \right) \frac{m_{H_2O}}{m_{mix}} = \left(\frac{2M_H}{M_{H_2O}} \right) F_{H_2O} \quad F_O = \frac{m_O}{m_{mix}} = \left(\frac{2M_O}{M_{PuO_2}} \right) \frac{m_{PuO_2}}{m_{mix}} + \left(\frac{M_O}{M_{H_2O}} \right) \frac{m_{H_2O}}{m_{mix}}$$

$$F_O = \left(\frac{2M_O}{M_{PuO_2}} \right) F_{PuO_2} + \left(\frac{M_O}{M_{H_2O}} \right) F_{H_2O}$$

Consider $V_{mix} = 1.0$ cc. Then $m_{mix} = \rho_{mix} V_{mix} = 4.650$ g.

Now we have $V_{PuO_2} = 0.3490$ cm³, $V_{H_2O} = 0.651$ cm³ so that

$$m_{PuO_2} = (0.3490)(11.46) = 3.9995 \text{ g} \quad m_{H_2O} = (0.6510)(1.0) = 0.6510 \text{ g}$$

$$F_{PuO_2} = \frac{m_{PuO_2}}{m_{mix}} = \frac{3.9995}{4.650} = 0.860 \quad F_{H_2O} = \frac{m_{H_2O}}{m_{mix}} = \frac{0.651}{4.650} = 0.140$$

$$\left(\frac{2M_O}{M_{PuO_2}} \right) = \frac{2(15.9995)}{271.0512} = 0.118055 \quad \left(\frac{M_O}{M_{H_2O}} \right) = \frac{15.9995}{18.01514} = 0.888114$$

$$F_O = \left(\frac{2M_O}{M_{PuO_2}} \right) F_{PuO_2} + \left(\frac{M_O}{M_{H_2O}} \right) F_{H_2O} = (0.118055)(0.86) + (0.888114)(0.14) = 0.22586$$

$$F_{Pu} = \left(\frac{239.0522}{271.0512} \right) F_{PuO_2} = 0.75848 \quad F_H = \left(\frac{2(1.00782)}{18.01514} \right) F_{H_2O} = 0.01566$$

$$n_{Pu} (@/B \times cm) = \frac{m_{Pu} N_A}{V_{mix} M_{Pu}} \times 10^{-24} = \frac{1}{V_{mix}} \left(\frac{M_{Pu}}{M_{PuO_2}} \right) \frac{m_{PuO_2}}{M_{Pu}} N_A \times 10^{-24} = \frac{m_{PuO_2}}{V_{mix}} \left(\frac{N_A}{M_{PuO_2}} \right) \times 10^{-24}$$

or $n_{Pu} (@/B \times cm) = 8.8859 \times 10^{-3}$

$$n_H (@/B \times cm) = \frac{1}{V_{mix}} \left(\frac{2M_H}{M_{H_2O}} \right) \frac{m_{H_2O}}{M_H} N_A \times 10^{-24} = 4.3523 \times 10^{-2}$$

$$n_O (@/B \times cm) = \frac{F_O \rho_{mix}}{M_O} N_A \times 10^{-24} = 3.9537 \times 10^{-2}$$

$$n_T = n_O + n_H + n_{Pu} = 9.19459 \times 10^{-2} \quad @/B \times cm$$

$$f_o = \frac{n_o}{n_T} = 4.3000 \times 10^{-1}$$

$$f_H = \frac{n_H}{n_T} = 4.7335 \times 10^{-1}$$

$$f_{Pu} = \frac{n_{Pu}}{n_T} = 9.6650 \times 10^{-2} .$$

Appendix C

Supplemental Results

Appendix C

Supplemental Results

Table C-1. 2.82-cm-thick slab containing 1.948 g/cm³ water saturated PuO₂, surrounded by water saturated soil.

Case Name	Slab Dimensions (ft)	Mass of Pu in Slab (kg)	$k_{\text{eff}} \pm 1\sigma$	$k_{\text{eff}} + 2\sigma$
TD389	2 × 2	20.41	0.8798 ± 0.0016	0.8830
TD381	4 × 4	81.66	0.9576 ± 0.0017	0.9610
TD382	6 × 6	183.73	0.9758 ± 0.0017	0.9792
TD386	10 × 10	510.35	0.9904 ± 0.0018	0.9940
TD388	12 × 12	734.90	0.9930 ± 0.0019	0.9968
TD059	Inf. × Inf.	—	0.9976 ± 0.0018	1.0012

Table C-2. 4.42-cm-thick slab containing 1.948 g/cm³ PuO₂ in water saturated soil, surrounded by water saturated soil.

Case Name	Slab Dimensions (ft)	Mass of Pu in Slab (kg)	$k_{\text{eff}} \pm 1\sigma$	$k_{\text{eff}} + 2\sigma$
TD399	2 × 2	32.00	0.8579 ± 0.0017	0.8613
TD391	4 × 4	127.99	0.9499 ± 0.0017	0.9533
TD392	6 × 6	287.97	0.9730 ± 0.0017	0.9764
TD394	8 × 8	511.94	0.9851 ± 0.0016	0.9883
TD396	10 × 10	799.91	0.9869 ± 0.0017	0.9903
TD398	12 × 12	1,151.87	0.9920 ± 0.0018	0.9956
TD317	Inf. × Inf.	—	0.9956 ± 0.0016	0.9988

Appendix D

Typical MCNP Input Listings

Appendix D

Typical MCNP Input Listings

D-1. MCNP LISTING FOR 10.0-CM RADIUS SPHERE OF WATER SATURATED PLUTONIUM DIOXIDE (1.948 G/CC), SURROUNDED BY WATER SATURATED SOIL.

```

ISTD112  10.0cm rad, 83%porous PU02 (1.948g/cc)
1  3  9.60705-02 -1          imp:n=1  $ Wet PuO2
2  1  -1.83          1  -2    imp:n=1  $ soil
3  0                      2    imp:n=0  $ Elsewhere

c  Surface Cards
1  so  10.0
2  so  243.84

c  Clean saturated soil
m1  14000.50c  1.0034-02  13027.50c  2.2387-03
    26000.55c  5.1263-04  20000.50c  6.3196-04
    19000.50c  6.1130-04  12000.50c  4.1102-04
    11023.50c  4.2591-04  22000.50c  8.2004-05
    25055.50c  1.1108-05
    5011.50c   1.3779-05  1001.50c  2.6742-02
    8016.50c   3.9335-02

mt1  lwtr.01t
c  Water in drum
m2  1001.50c  2          8016.50c  1
mt2  lwtr.01t
c  Wet PuO2
m3  1001.50c  5.53902-02  8016.50c  3.63519-02
    94239.50c  4.32843-03

mt3  lwtr.01t
kcode 1800 1.0 25 165
ksrc 0. 0. 0.066 0. 0. 0.1 0. 0. 0.2
print

```

D-2. MCNP LISTING FOR 10.5-CM RADIUS SPHERE OF PLUTONIUM DIOXIDE (4.584 G/CC) AND WATER SATURATED SOIL, SURROUNDED BY WATER SATURATED SOIL.

```

ISTD204  10.5cm rad, 60%porous PU02 (4.584g/cc),in soil
1  3  7.91833-02 -1          imp:n=1  $ PuO2 in soil
2  1  -1.83          1  -2    imp:n=1  $ soil
3  0                      2    imp:n=0  $ Elsewhere

c  Surface Cards
1  so  10.5
2  so  243.84

c  Clean saturated soil
m1  14000.50c  1.0034-02  13027.50c  2.2387-03
    26000.55c  5.1263-04  20000.50c  6.3196-04
    19000.50c  6.1130-04  12000.50c  4.1102-04
    11023.50c  4.2591-04  22000.50c  8.2004-05
    25055.50c  1.1108-05
    5011.50c   1.3779-05  1001.50c  2.6742-02
    8016.50c   3.9335-02

mt1  lwtr.01t
c  Water in drum
m2  1001.50c  2          8016.50c  1
mt2  lwtr.01t
c  PuO2 in soil

```

```

m3 14000.50c 6.0204-03 13027.50c 1.3432-03
    26000.55c 3.0758-04 20000.50c 3.7918-04
    19000.50c 3.6678-04 12000.50c 2.4661-04
    11023.50c 2.5555-04 22000.50c 4.9202-05
    25055.50c 6.6648-06
    5011.50c 8.2674-06 1001.50c 1.6045-02
    8016.50c 4.3970-02
    94239.50c 1.01845-02
mt3 lwtr.01t
kcode 1800 1.0 25 165
ksrc 0. 0. 0.066 0. 0. 0.1 0. 0. 0.2
print

```

D-3. MCNP LISTING FOR 4.5-CM-THICK SLAB OF WATER SATURATED PLUTONIUM DIOXIDE (1.948 G/CC) IN 55-GALLON DRUM, SURROUNDED BY WATER SATURATED SOIL.

```

ISTD182 4.5cm thick, 83%porous PU02 (1.948g/cc),in drum
1 3 9.60705-02 -3 2 -5 imp:n=1 $ Wet Pu02
2 1 -1.83 (3:-2:5) -4 1 -6 imp:n=1 $ soil
3 0 (4:-1:6) imp:n=0 $ Elsewhere

```

```

c Surface cards
1 pz -121.92
2 pz 0.0
3 pz 4.5
4 pz 731.52
5 cz 28.575
6 cz 150.5

```

```

c Clean saturated soil
m1 14000.50c 1.0034-02 13027.50c 2.2387-03
    26000.55c 5.1263-04 20000.50c 6.3196-04
    19000.50c 6.1130-04 12000.50c 4.1102-04
    11023.50c 4.2591-04 22000.50c 8.2004-05
    25055.50c 1.1108-05
    5011.50c 1.3779-05 1001.50c 2.6742-02
    8016.50c 3.9335-02

```

```

mt1 lwtr.01t
c water in drum
m2 1001.50c 2 8016.50c 1
mt2 lwtr.01t
c wet Pu02
m3 1001.50c 5.53902-02 8016.50c 3.63519-02
    94239.50c 4.32843-03
mt3 lwtr.01t
kcode 1800 1.0 25 165
ksrc 0. 0. 0.066 0. 0. 0.1 0. 0. 0.2
print

```

D-4. MCNP LISTING FOR 8.5-CM-THICK SLAB OF PLUTONIUM DIOXIDE (1.948 G/CC) AND WATER SATURATED SOIL IN 55-GALLON DRUM, SURROUNDED BY WATER SATURATED SOIL.

```

ISTD375 8.0cm thick, 83%porous PU02 (1.948g/cc),in soil drum
1 3 8.02563-02 -3 2 -5 imp:n=1 $ Pu02 in soil
2 1 -1.83 (3:-2:5) -4 1 -6 imp:n=1 $ soil
3 0 (4:-1:6) imp:n=0 $ Elsewhere

```

```

c Surface Cards
1 pz -121.92
2 pz 0.0
3 pz 8.0

```

```

4 pz 731.52
5 cz 28.575
6 cz 150.5

c Clean saturated soil
m1 14000.50c 1.0034-02 13027.50c 2.2387-03
    26000.55c 5.1263-04 20000.50c 6.3196-04
    19000.50c 6.1130-04 12000.50c 4.1102-04
    11023.50c 4.2591-04 22000.50c 8.2004-05
    25055.50c 1.1108-05
    5011.50c 1.3779-05 1001.50c 2.6742-02
    8016.50c 3.9335-02
mt1 lwtr.01t
c water in drum
m2 1001.50c 2 8016.50c 1
mt2 lwtr.01t
c Wet PuO2
m3 14000.50c 8.3282-03 13027.50c 1.8581-03
    26000.55c 4.2548-04 20000.50c 5.2453-04
    19000.50c 5.0738-04 12000.50c 3.4115-04
    11023.50c 3.5351-04 22000.50c 6.8063-05
    25055.50c 9.2196-06
    5011.50c 1.1437-05 1001.50c 2.2196-02
    8016.50c 4.1305-02
    94239.50c 4.32843-03
mt3 lwtr.01t
kcode 1800 1.0 25 165
ksrc 0. 0. 0.066 0. 0. 0.1 0. 0. 0.2
print

```

D-5. MCNP LISTING FOR 2.82-CM-THICK INFINITE SLAB OF WATER SATURATED PLUTONIUM DIOXIDE (1.948 G/CC), SURROUNDED BY WATER SATURATED SOIL.

```

ISTD059 2.82cm thick, 83%porous PUO2 (1.948g/cc)
1 1 -1.83 -2 1 -6 5 -8 7 imp:n=1 $ soil
2 3 9.60705-02 -3 2 -6 5 -8 7 imp:n=1 $ Wet PuO2
3 1 -1.83 -4 3 -6 5 -8 7 imp:n=1 $ soil
4 0 -1:4:6:-5:8:-7 imp:n=0 $ Elsewhere

```

```

c Surface Cards
1 pz -121.92
2 pz 0.0
3 pz 2.82
4 pz 731.52
*5 px -25.0
*6 px 25.0
*7 py -25.0
*8 py 25.0

```

```

c Clean saturated soil
m1 14000.50c 1.0034-02 13027.50c 2.2387-03
    26000.55c 5.1263-04 20000.50c 6.3196-04
    19000.50c 6.1130-04 12000.50c 4.1102-04
    11023.50c 4.2591-04 22000.50c 8.2004-05
    25055.50c 1.1108-05
    5011.50c 1.3779-05 1001.50c 2.6742-02
    8016.50c 3.9335-02
mt1 lwtr.01t
c water in drum
m2 1001.50c 2 8016.50c 1
mt2 lwtr.01t
c Wet PuO2
m3 1001.50c 5.53902-02 8016.50c 3.63519-02
    94239.50c 4.32843-03
mt3 lwtr.01t
kcode 1800 1.0 25 165

```



```
ksrc 0. 0. 0.066    0. 0. 0.1    0. 0. 0.2
print
```

D-6. MCNP LISTING FOR 4.42-CM-THICK INFINITE SLAB OF PLUTONIUM DIOXIDE (1.948 G/CC) AND WATER SATURATED SOIL, SURROUNDED BY WATER SATURATED SOIL.

```
ISTD317 4.42cm thick, 83%porous PU02 (1.948g/cc),in soil
1 1 -1.83 -2 1 -6 5 -8 7 imp:n=1 $ soil
2 3 8.02563-02 -3 2 -6 5 -8 7 imp:n=1 $ PuO2 in soil
3 1 -1.83 -4 3 -6 5 -8 7 imp:n=1 $ soil
4 0 -1:4:6:-5:8:-7 imp:n=0 $ Elsewhere
```

c Surface Cards

```
1 pz -121.92
2 pz 0.0
3 pz 4.42
4 pz 731.52
*5 px -25.0
*6 px 25.0
*7 py -25.0
*8 py 25.0
```

c Clean saturated soil

```
m1 14000.50c 1.0034-02 13027.50c 2.2387-03
    26000.55c 5.1263-04 20000.50c 6.3196-04
    19000.50c 6.1130-04 12000.50c 4.1102-04
    11023.50c 4.2591-04 22000.50c 8.2004-05
    25055.50c 1.1108-05
    5011.50c 1.3779-05 1001.50c 2.6742-02
    8016.50c 3.9335-02
```

mt1 lwtr.01t

c Water in drum

```
m2 1001.50c 2 8016.50c 1
```

mt2 lwtr.01t

c Wet PuO2

```
m3 14000.50c 8.3282-03 13027.50c 1.8581-03
    26000.55c 4.2548-04 20000.50c 5.2453-04
    19000.50c 5.0738-04 12000.50c 3.4115-04
    11023.50c 3.5351-04 22000.50c 6.8063-05
    25055.50c 9.2196-06
    5011.50c 1.1437-05 1001.50c 2.2196-02
    8016.50c 4.1305-02
    94239.50c 4.32843-03
```

mt3 lwtr.01t

kcode 1800 1.0 25 165

```
ksrc 0. 0. 0.066    0. 0. 0.1    0. 0. 0.2
```

print